

Criticality Safety Evaluation for In Situ Vitrification Processing (ISV) at the Radioactive Waste Management Complex at INEEL

Paul Sentieri

April 2003

Idaho National Engineering and Environmental Laboratory Bechtel BWXT Idaho, LLC

INEEL/EXT-03-00207 Revision 0

Project No: 023378

Criticality Safety Evaluation for In Situ Vitrification Processing (ISV) at the Radioactive Waste Management Complex at INEEL

Paul Sentieri

April 2003

Idaho National Engineering and Environmental Laboratory Environmental Restoration Program Idaho Falls, Idaho 83415

Prepared for the
U.S. Department of Energy
Assistant Secretary for Environmental Management
Under DOE Idaho Operations Office
Contract DE-AC07-99ID13727

Criticality Safety Evaluation for In Situ Vitrification Processing at the Radioactive Waste Management Complex at INEEL

INEEL/EXT-03-00207 Revision 0

Author	4/29/03
Paul J. Sentieri Criticality Safety Engineer	Date
Checked by	
Mark Heeley	4/29/03
Mark N. Neeley	
Criticality Safety Engineer	Date
Approved by	
ful My	4/29/03
Brandt G. Meagher	/ / Date
Operable Unit 7-13/14 Project Manager	
	4/30/03
Doug S. Swanson	/ / Date
Safety Analysis	
- Have	4/30/03
J. Todd Taylor	Date
Criticality Safety Department Manager	

ABSTRACT

The purpose of this document is to address the potential for a criticality in the Subsurface Disposal Area (SDA) due to the proposed in situ vitrification (ISV) process. A criticality safety study was performed to address issues relating to postulated criticality scenarios in the SDA for Operable Unit 7-13/14 in the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory (INEEL).

Based on the results of this study, a criticality resulting from the application of the ISV process is not credible with the expected fissile masses and waste forms in the SDA.

The author would like to acknowledge what an important role the initial work, completed by Roger Gathers of M. L. Chew and Associates, played in the development of this report. Mr. Gathers completed the initial scenario development, computational modeling, number density calculations, and other calculations used in support of this evaluation. Mr. Gathers performed this effort in a subcontracting role for the INEEL. His initial work was used as the basis for this evaluation.



EXECUTIVE SUMMARY

The Subsurface Disposal Area (SDA) is that portion of the Radioactive Waste Management Complex (RWMC) that was established in 1952 as a disposal site for solid low-level radioactive waste. Transuranic waste was received from the Rocky Flats Plant and disposed of in the Subsurface Disposal Area from 1954 to 1970. This study examines criticality safety issues associated with the use of in situ vitrification (ISV) as a means of immobilizing the buried transuranic waste in the SDA. For the safety studies it was decided to only analyze ²³⁹Pu, since it is by far the most reactive and abundant fissile material reported to be disposed of.

Various configurations were evaluated to determine if any criticality concerns arise in conjunction with treating the buried transuranic waste contained in the SDA with ISV. This evaluation consisted of three phases. The first phase was to consider criticality scenarios during the initial application of ISV. The second phase consisted of evaluating the final configurations as a result of the application of ISV. The third phase was to address ancillary issues relating to ISV and criticality safety.

During the initial application of ISV, the fissile-bearing waste within the SDA is subjected to various physical and mechanical processes. These processes include the melting of metals, the oxidation of metals, the melting of plastics that could entrain fissile material, subsidence in the waste zone, creation of convective currents in the melt zones that will disperse the fissile material, and the eventual formation of vitrified waste materials.

These processes led to the development of the scenarios and final configurations that were evaluated. Two scenarios were postulated that could lead to unsafe conditions. The first is the melting and eventual reconfiguration of fissile material in the form of molten metal in an unsafe configuration. The second is the formation of an unsafe condition, due to fissile material becoming entrained in or mixed with plastic and forming an unsafe condition prior to the eventual removal of the plastics due to the ISV process.

The evaluation of the final configuration consists of three parts. The first part is the determination of the fissile concentration necessary to achieve an unsafe configuration in a vitrified soil block. The second part is the comparison of the reactivity between vitrified soil and water-saturated soil. This evaluation showed that once the waste matrices are vitrified, a higher concentration is necessary to achieve an unsafe condition. The third part evaluated was the localized effects of an overloaded drum becoming vitrified in the presence of other fissile bearing waste materials.

Additionally, this evaluation addressed the effects of a postulated melt expulsion, re-entry of water into the final configuration, and whether it is credible to form a critical system in the off gas collection system. Some of the postulated configurations were evaluated by qualitative means, while other configurations were addressed through computational modeling. Based on the results of this study, a criticality due to the application of the ISV process is not credible with the expected fissile masses and waste forms in the SDA.

CONTENTS

ABS	TRAC	T	iii
EXE	CUTIV	VE SUMMARY	v
1.	INTI	RODUCTION	1
	1.1	Description of the RWMC	1
	1.2	Criticality Safety Background Information and Definitions	1
2.	DES	CRIPTION	3
	2.1	In Situ Vitrification	3
3.	REQ	UIREMENTS DOCUMENTATION	4
4.	MET	HODOLOGY	4
5.	DISC	CUSSION OF CONTINGENCIES	6
6.	EVA	LUATION & RESULTS	7
	6.1	Melting and Concentration of ²³⁹ Pu or ²³⁵ U Metal	8
	6.2	Moderation of Pu by Mixing with Organic Materials such as Polyethylene	9
	6.3	Distribution of Plutonium in a Vitrified Matrix	11
	6.4	Comparison between PuO ₂ distributed in Vitrified Soil or Water Saturated Soil	15
	6.5	Arrays of Drums Containing Pu distributed in Vitrified Soil and Saturated Soil	16
	6.6	Melt Expulsion	19
	6.7	Flooding and/or Water Re-entry	20
	6.8	Collection of Fissile Materials in the Off-Gas System	21
7.	DES	IGN FEATURES AND ADMINISTRATIVE CONTROLS & LIMITS	22
8.	SUM	IMARY AND CONCLUSIONS	22
9.	REF.	ERENCES	24
		—Materials and Compositions	
		B—Supporting Calculations and Methods C—Composition Calculational Spreadsheets	
		— Composition Calculational Spreadsheets — Typical MCNP Input Listings	

FIGURES

1.	Calculations of k_{eff} for a slab of 239 PuO ₂ mixed with polyethylene located at the bottom of a drum surrounded with vitrified soil. The remaining volume of the drum is filled with full density polyethylene	11
2.	$k_{\rm eff}$ versus Pu concentration for an infinite slab of vitrified soil, mixed with PuO ₂ for two values of slab thickness and varied Pu concentrations. The slab is bounded by 4 ft of vitrified clean soil below, and by 6 ft of dry, porous, clean soil above. The four sets of curves are for varied SiO_2 content in the soil.	14
3.	Calculations of $k_{\rm eff}$ for spheres of $^{239}\text{PuO}_2$ mixed with either vitrified soil or 40% porous water-saturated soil, and reflected by 40% porous water-saturated soil	17
4.	Graphical Representation of Results from Tables 7 and 8	20
	TABLES	
1.	Listing of RWMC waste matrix designations	3
2.	Calculated results for the plutonium experiments	5
3.	Calculated results for the PuO _{2/p} olystyrene experiments	5
4.	Infinite horizontal slab of vitrified soil of various concentrations of SiO ₂ mixed with various concentrations of Pu as oxide	12
5.	Finite slabs of vitrified soil, 2.5 g/l concentration of Pu as oxide. The slab is bounded by vitrified soil, 4-ft-thick below, and by dry porous soil 6 ft thick above. The slab is 14 ft thick. No credit is taken for ¹⁰ B.	14
6.	Infinite slab of vitrified soil with 10 wt% C added, and with various concentrations of Pu as oxide. The slab is bounded by vitrified soil 4 ft thick below, and by dry porous soil 6 ft thick above. The slab is either 10 ft thick or 14 ft thick. No credit is taken for ¹⁰ B.	15
7.	MCNP calculations of $k_{\rm eff}$ versus Pu concentration for 1200 g Pu as oxide and mixed with either vitrified soil or 40% porous saturated soil, surrounded by saturated soil. No 10 B present	16
8.	$11 \times 11 \times 3$ array of drums, fissile material mixed in vitrified matrix, nominal loading per drum 200 g Pu. Four adjacent drums near the center of the array are abnormal (1200 g Pu as oxide mixed with vitrified soil, and the other three contain 300 g Pu as oxide mixed with vitrified soil). The abnormal spheres are clustered together. The surrounding medium is either vitrified soil or 40% porous, water-saturated soil	18
9.	$11 \times 11 \times 3$ array of drums fissile material mixed in saturated soil, nominal loading per drum 200 g Pu. Four adjacent drums near the center of the array are abnormal (1200 g Pu as oxide mixed with saturated soil, and the other three contain 300 g Pu as oxide mixed with saturated soil). The abnormal spheres are clustered together. The surrounding medium is 40% porous, water-saturated soil	19

Criticality Safety Evaluation for In Situ Vitrification Processing at the Radioactive Waste Management Complex at INEEL

1. INTRODUCTION

The purpose of this document is to address the potential for a criticality in the Subsurface Disposal Area (SDA) resulting from the application of the in situ vitrification (ISV) process.

The conventional ISV process works by melting soil and waste in place using electricity applied between pairs of graphite electrodes. The electrodes are inserted in a square or rectangular configuration up to 26 ft apart and 1-2 ft into the ground. A highly conductive mixture of graphite and glass-forming compounds is placed between the electrodes for startup purposes. When an electric potential is applied to the electrodes, current flows through the starter path, which heats up and causes the surrounding soil to melt. Once the soil is molten, it too becomes electrically conducting. Continued application of electricity results in joule heating (heating due to electrical resistance) within the molten soil between the electrodes. After the melt is fully established, the melt zone grows steadily downward and outward through the contaminated media, as the graphite electrodes are gravity-fed downward into the melt. Electrical power is delivered to a transformer at the site. As the melt zone increases in size, the resistance of the electrical load decreases and the power factor is adjusted by using taps on the transformer to maintain efficiency in the heating process. For ISV activity, a modified version called planar ISV will be used. Melting is established in multiple planes. The melt(s) grow outward and downward from each plane, eventually coalescing together.

1.1 Description of the RWMC

The Radioactive Waste Management Complex (RWMC) was established in the early 1950s as a disposal site for solid low-level waste generated by Idaho National Engineering and Environmental Laboratory (INEEL) operations. The RWMC has been designated as Waste Area Group (WAG) 7 and is subdivided into 14 Operable units (OUs). Within the RWMC is the Subsurface Disposal Area (SDA), where radioactive waste materials have been buried in underground pits, trenches, soil vault rows, etc. Transuranic waste was disposed of in the SDA from 1952 to 1970 and was received from Rocky Flats Plant (RFP) for disposal in the SDA from 1954 to 1970. The RFP is a U.S. Department of Energy (DOE)-owned facility located west of Denver, Colorado, and was used primarily for the production of components for nuclear weapons.

The Operable unit (OU) 7-13/14 technology demonstrations for ISV will provide sufficient data to evaluate the technologies for buried waste treatments for long-term in situ disposal at the SDA.

1.2 Criticality Safety Background Information and Definitions

Waste at the RWMC contains fissile isotopes. The fissile isotopes of concern are 239 Pu from weapons-grade plutonium (94-wt% 239 Pu) and 235 U from highly enriched uranium (93-wt% 235 U). Much of the fissile material is in oxide, nitrate, or hydrated-oxide form. Criticality is difficult to obtain and requires special conditions and materials. It may be defined as "the attainment of conditions such that fissile material will sustain a chain reaction (Cember 1985)." Criticality is determined by reactivity. The term "reactivity" refers to the deviation of a system from a critical value of k_{eff} =1, where k_{eff} is the neutron multiplication factor for a system. A more reactive system has greater neutron multiplication. If

the neutron multiplication is such that the system is just self-sustaining, it is critical. A supercritical system has a value of $k_{\rm eff}$ greater than 1, while a sub critical system has a $k_{\rm eff}$ value less than 1. The factors that affect reactivity are reflection, moderation, geometry, fissile mass, and fissile mass concentration.

A reflector surrounds the fissile material and reflects neutrons back into the fissile region. Reflectors include water, paraffin, beryllium, graphite, concrete, and thick metal. A moderator is usually intimately mixed with the fissile material. It slows neutrons so that they are more likely to react with the fissile isotopes and cause fission. Many reflectors are also good moderators. Hydrogen is the most common and effective moderator, particularly as found in water, polyethylene, or in oils. Hydrogen is an effective moderating atom because it has almost the same mass as a neutron enabling almost complete momentum transfer in a single collision. Oil and polyethylene are better moderators than water because they have greater hydrogen density. Beryllium and graphite are also good moderators, but require larger volumes for criticality. Beryllium is actually more effective as a reflector than as a moderator. Each system has an optimally reflected, moderated state where its reactivity is greatest.

The geometry of the fissile material affects the system reactivity. As the ratio of volume to surface area increases for a given volume, neutron leakage decreases and reactivity increases (Paxton & Pruvost 1986). The optimum geometry is thus usually a sphere that has maximum volume and minimum surface area. The type of fissile isotope affects the system reactivity. Odd numbered isotopes usually fission due to the absorption of thermal neutrons more readily than even numbered isotopes. Even numbered isotopes more readily fission due to fast spectral neutrons. Thus, ²³⁵U and ²³⁹Pu are much more fissionable than ²³⁸U and ²⁴⁰U. The concentration of the fissile isotope is important to the system reactivity. If the fissile material is too dilute, criticality cannot be achieved.

Moderation and reflection usually work together since most moderators also act as reflectors. Much smaller masses of fissile isotopes dissolved with water must be maintained to keep a non-critical configuration. A fissile material solution in a moderator, such as water, has a smaller critical mass. The minimum critical mass in water for each of the fissile isotopes corresponds to an optimally moderated and homogeneous mixture of the metal isotope and water, in a spherical configuration with full water reflection. These values are specified as the "solution critical mass." The critical mass of the solid metal is more than ten times that for the isotope in solution. Moist-oxide critical masses are significantly greater than those for solutions and metals. Deviations from the ideal conditions increase the critical mass.

Approximately 20,000 drums from aboveground storage operations at the RWMC have been assayed for fissile content utilizing several nondestructive assay methods. Results from the assaying of these 20,000 drums show that no drum has a fissile loading in excess of 380 g. About 100 drums have fissile loading in excess of 200 g. This indicates that localized concentration of a large quantity of buried fissile material is not likely to be encountered within the SDA.

Initial probing data within one portion of the SDA indicated one possible location of higher fissile mass. The conservative estimates for the fissile mass ranged from an unlikely probability of 547 g to an extremely unlikely probability of 2,221 g of fissile material being present (Peatross 2003). This area is located in OU 7-10 and will be excavated and quantified prior to the application of ISV in any fashion within the SDA. The data can then be analyzed relating to whether or not the actual fissile content present is representative of the estimates.

The RWMC assigns a content code to each waste container. These are based on the process used for the waste. "Sludges" make up the predominant mass and volume of the waste. There are three general process "sludge" type wastes, inorganic (741-742), organic (743-744), and salt (745). The other waste is generally debris (concrete/asphalt), metal, and trash (combustibles). For criticality safety purposes, these content codes are grouped into eight waste matrices. A waste matrix can cover a range of materials.

Table 1 lists the RWMC criticality waste matrix designations and gives some examples of waste covered by each matrix.

Table 1. Listing of RWMC waste matrix designations.

General Classification	Waste Matrix	Examples of Typical Waste
Organic Sludge	Oil and clay	Resins and combustibles
Combustible	Cellulose	Benelex, Plexiglas, cemented insulation and filter media
Debris	Brick	Fire brick – scarfed, coarse, pulverized
Debris, Inorganic sludge	Concrete	Cemented and un-cemented sludges
Salts	Salt	Evaporated, molten, Gibson, direct oxide reduction salts
Metal	Metal	Non-combustibles, non-compressibles, tantalum, lead
Debris	Glass/slag	Glass bottles, crucibles and molds, dirt, ceramic crucible

2. DESCRIPTION

2.1 In Situ Vitrification

The conventional ISV process works by melting soil and waste in place using electricity applied between pairs of graphite electrodes. The electrodes are inserted in a square or rectangular configuration up to 26 ft apart and 1–2 ft into the ground. A highly conductive mixture of graphite and glass-forming compounds is placed between the electrodes for startup purposes. When an electric potential is applied to the electrodes, current flows through the starter path, which heats up and causes the surrounding soil to melt. Once the soil is molten, it too becomes electrically conducting. Continued application of electricity results in joule heating within the molten soil between the electrodes. After the melt is fully established, the melt zone grows steadily downward and outward through the contaminated media, as the graphite electrodes are gravity-fed downward into the melt. Electrical power is delivered to a transformer at the site. As the melt zone increases in size, the resistance of the electrical load decreases and the power factor is adjusted by using taps on the transformer to maintain efficiency in the heating process. For the ISV activity, a modified version called planar ISV will be used. Melting is established in multiple planes. The melt(s) grow outward and downward from each plane, eventually coalescing together.

During ISV processing, nonvolatile metals and radio nuclides are chemically incorporated as oxides into the vitrified product. Volatile metals (e.g., mercury) are vaporized and removed by an off-gas treatment system. Organic contaminants are typically destroyed by pyrolysis in the ground. The pyrolysis products travel to the ground surface where they are oxidized in the off-gas hood. Vapors from the off-gas hood are then processed by the off-gas treatment system before being discharged to the atmosphere. The highly-reducing nature of the ISV melt may result in some metals (e.g., iron) being reduced if sufficient quantities exist in the soil (<15-20 wt%) or staying in a reduced form, and settling to the bottom of the melt as a separate phase. Vitrification work performed on high-level waste has demonstrated that some radio nuclides, for example uranium and plutonium (two of the contaminants of potential concern for OU 7-13/14), will not be reduced due to their high oxidation potentials and therefore will remain in the glass phase (Buelt, et al 1987.). The ISV process utilizes very high temperatures (2000°C) and produces strong convective currents in the melt. As a result, the buried materials are generally made into a more homogeneous mixture. This is beneficial in that dispersal of the fissile materials will decrease reactivity.

The ISV process will produce a vitreous rock-like material that is virtually free of organic material and has a physical strength approximately 10 times greater than nonreinforced concrete. The vitrified

product is extremely resistant to leaching and will typically pass even the most stringent leach tests. The life expectancy of the vitrified product is expected to be similar to naturally occurring obsidian or basalt, which has a life expectancy measured in millions of years when exposed to the natural environment (Buelt et al).

Further description of the ISV process can be found in references Farnsworth et al. (1999), and Callow (1991).

3. REQUIREMENTS DOCUMENTATION

PRD-112 allows a maximum $k_{eff} + 2\sigma$ of 0.95 under single accident conditions (INEEL 1998). No unique requirements are applicable to this evaluation.

4. METHODOLOGY

Calculations were performed using the MCNP, version 4B2, computer code system (RSIC 1997). MCNP is a Monte Carlo transport code used to determine $k_{\rm eff}$ for systems containing fissionable material. The cross-section libraries used for this analysis contained the "point-wise" or "continuous-energy" cross sections, although MCNP can be used with multigroup cross sections also.

The analyzed system contained in this report consisted of plutonium dispersed in various waste matrices including vitrified soil, saturated soil, and polyethylene. The geometry of the evaluated systems compromised waste materials and plutonium in cylindrical form (drums), spherical form (optimized systems), and rectangular form (slabs) of both finite and infinite extents in the horizontal directions.

No critical experiments exist that exactly match the types of systems evaluated. However, modeling critical experiments that encompass the evaluated parameters can validate the various models. These parameters include material composition, moderation conditions, reflection conditions, and spectral neutron energy ranges.

Validation for these calculations requires experiments that consist of moderated plutonium systems and plutonium combined with silicon.

A separate report was completed that evaluated critical plutonium/silicon configurations.^a Experiments consisting of plutonium fuel rods, intermixed in a triangular lattice with silicon/dioxide rods, were performed in Obninsk, Russia in 1998 and 1999. A detailed description of the critical configurations can be found in Tsiboulia et al. (2000).

A brief description of the experiments follows. Ten different types of rods were used in the plutonium experiments. Each of the rods consisted of a stack of various discs or pellets of various materials. These materials included plutonium metal (canned in stainless steel), silica pellets, polyethylene pellets, stainless steel pellets, and boron carbide pellets. Each of the 10 different rods contained a combination of these pellets in a stacked configuration. The rods were then combined to create a critical system. The fuel tubes were arranged in a hexagonal array with a 5.1-cm pitch.

The experiments were modeled as described previously. The calculated results for the experiments, using the ENDF/B-V cross section library, are provided in Table 2. The H/Pu ratio and Si/Pu ratio for the

4

a. This Report, completed in 2002 by J.W. Nielsen, concerned validation of uranium and plutonium silicon dioxide experiments.

experiments is also presented in the table. The H/Pu ratio varied from 0 to 35 while the Si/Pu ratio varied from 23 to 42. The calculated neutron energy spectrum for these experiments indicates that the energy of the neutrons causing fission is primarily in the intermediate (0.625 eV to 100 keV) to fast (more than 100 keV) range. The average calculated k_{eff} for these experiments is 1.0070 ± 0.0003 .

Table 2. Calculated results for the plutonium experiments.

Case Name	H/Pu	Si/Pu	$k_{\rm eff}\pm\sigma$
BFS-81/1	0	23.4	1.0001 ± 0.0006
BFS-81/1A	0	23.4	0.9987 ± 0.0008
BFS-81/2	2.8	23.4	1.0055 ± 0.0008
BFS-81/3	5.6	23.4	1.0089 ± 0.0008
BFS-81/4	35.2	41.6	1.0178 ± 0.0008
BFS-81/5	35.2	41.6	1.0164 ± 0.0008
Average: $k_{avg} = \sum (k_i/\sigma_i^2)/\sum (1/\sigma_i^2)$,			1.0070 ± 0.0003

a. ICSEBP 2000, "International Handbook of Evaluated Criticality Safety Benchmark Experiments", NEA Nuclear Science Committee, Nuclear Energy Agency, NEA/ANSC/DOC(95)03, September 2000

The last set of evaluated cases consisted of PuO₂/polystyrene and were reflected by plexiglass. Experiments were performed at Hanford between 1963 and 1970. The experiments consisted of cubes of PuO₂/polystyrene reflected by plexiglass plates. Twenty-nine experiments were performed with various configurations, concentrations of plutonium, and plutonium enrichments.

The cubes were approximately $2\times2\times2$ in. and were stacked on a split-table critical assembly. The two halves of the assembly were brought together and the neutron multiplication was determined using proportional counters. Some of the cubes were cut in the axial direction to allow flexibility in obtaining a critical height. The final critical configuration consists of a rectangular block of PuO_2 -polystyrene reflected on all six sides by plexiglass. The H/Pu ratios ranged from 5.87–65.4 with the C/Pu ratios varying 5.86–64.4. A more detailed description of these experiments can be found in an internal report (Nielsen 2003) that discusses validation of calculations containing HEU/graphite and Pu/polystyrene. The results from these cases can be found in Table 3.

As shown by the results of these validation experiments, no bias caused by calculational methodology is warranted.

Table 3. Calculated results for the PuO₂/polystyrene experiments.

	Case Name	$k_{\rm eff} \pm \sigma$	
Case 6		1.0170 ± 0.0009	
Case 7		1.0177 ± 0.0008	
Case 8		1.0173 ± 0.0007	
Case 9		1.0193 ± 0.0008	
Case 10		1.0285 ± 0.0010	
Case11		1.0270 ± 0.0010	

Table 3. (continued).

Case Name	$k_{eff}\pm\sigma$
Case 12	1.0247 ± 0.0010
Case 13	1.0233 ± 0.0009
Case 14	1.0275 ± 0.0010
Case 15	1.0256 ± 0.0009
Case 16	1.0214 ± 0.0010
Case 17	1.0045 ± 0.0009
Case 18	1.0088 ± 0.0008
Case 19	1.0051 ± 0.0007
Case 20	1.0056 ± 0.0008
Case 21	1.0072 ± 0.0009
Case 22	1.0101 ± 0.0008
Case 23	1.0054 ± 0.0009
Case 24	1.0054 ± 0.0008
Case 25	1.0069 ± 0.0017
Case 26	1.0081 ± 0.0009
Case 27	1.0086 ± 0.0008
Case 28	1.0091 ± 0.0009
Case 29	1.0110 ± 0.0010
Average: $k_{avg} = \sum (k_i/\sigma_i^2)/\sum (1/\sigma_i^2)$, $\sigma_{avg} = (1/\sum (1/\sigma_i^2))^{1/2}$	1.0138 ± 0.0002

a. ICSEBP 2000, "International Handbook of Evaluated Criticality Safety Benchmark Experiments", NEA Nuclear Science Committee, Nuclear Energy Agency, NEA/ANSC/DOC(95)03, September 2000

5. DISCUSSION OF CONTINGENCIES

An inadvertent criticality in the SDA caused by the ISV process is not possible.

In order to create a critical configuration with reasonable quantities of fissile material, various factors must be met. An unsafe mass of fissile material must be present. This fissile mass must be concentrated and in a favorable or optimal geometrical configuration. The system needs full reflection and must be free from diluent- or neutron-absorbing materials.

The fissile material in the SDA is dispersed at relatively low concentrations. If an area of fissile material existed with higher concentrations, the various factors above would need to be near-optimal in order to achieve an unsafe condition. Approximately 10.2 kg of moist $(1.5 \text{ wt}\% \text{ water}) \text{ PuO}_2$ is required to create an unsafe condition. This system consists of uniform concentration of fissile material in a small volume, which is free of diluent materials and fully reflected by an infinite perfect reflector. This is not the case in the SDA. These idealistic conditions do not exist in the SDA. The application of the ISV process will not create these conditions.

For more reasonable fissile masses, the optimal conditions are even more necessary to create an unsafe condition. These optimal conditions do not exist in the SDA. The application of the ISV process will create a melt that will disperse the fissile material and mix it with other waste forms, thus mixing diluent- or neutron-absorbing material intimately with the fissile material present. Additionally, the application of ISV will remove moderating materials and will create a vitrified matrix that prevents moderation from water, due to future water intrusion.

6. EVALUATION & RESULTS

For criticality to occur in the SDA (due to the ISV process), several unlikely concurrent parameters must exist: a) there must be sufficient fissile mass, b) the fissile mass must be at or near the optimum concentration, c) the fissile mass must be in a near optimal geometry, d) near optimal reflection must exist, and e) the fissile mass must be in a waste matrix that lacks diluent and neutronic absorber. The ISV process will remove and eventually exclude moderating materials from the vitrified matrix.

Various configurations were evaluated to determine if any criticality concerns arose in conjunction with using the ISV process to treat buried waste contained in the SDA. This evaluation consisted of three phases: 1) considering criticality scenarios during the initial application of the ISV process, 2) evaluating the final configurations as a result of the application of the ISV process, and 3) addressing ancillary issues relating to ISV and criticality safety. Each of these phases is further described in the remainder of this section.

During the initial application of the ISV process, the fissile-bearing waste within the SDA is subjected to various processes prior to the eventual vitrification of the waste matrices. These processes include the melting of metals, the oxidation of metals, the melting of plastics that could entrain fissile material, subsidence in the waste zone, creation of convective currents in the melt zones that will disperse the fissile material, and the eventual formation of vitrified waste materials.

These physical and mechanical processes lead to the development of the scenarios and final configurations that will be evaluated in the first phase. Two postulated scenarios that could lead to an unsafe condition will be evaluated. The first scenario is the melting and eventual reconfiguration of fissile material in the form of molten metal in an unsafe configuration. The second scenario is the formation of an unsafe condition due to fissile material becoming entrained in or mixed with plastic. During the scenario, this fissile material could form an unsafe condition, prior to the eventual removal of the plastics, due to the ISV process.

The second phase evaluates the final configuration. This second phase consists of three parts: 1) determining the fissile concentration necessary to achieve an unsafe configuration in a large vitrified soil block, 2) comparing the reactivity between vitrified soil and water-saturated soil (in order to show that once the waste matrices are vitrified, a higher concentration is necessary to achieve an unsafe condition), and 3) evaluating the localized effects of an overloaded drum becoming vitrified in the presence of other fissile-bearing waste materials.

The third phase of this evaluation will address the effects of a postulated melt expulsion, re-entry of water into the final configuration, and whether it is credible, from a criticality safety standpoint, to form an unsafe condition in the off-gas collection system.

Some of the postulated configurations were evaluated by qualitative means while other configurations were addressed through computational modeling. A more detailed assessment of these postulated configurations and the evaluation of the final configuration will now be presented.

6.1 Melting and Concentration of ²³⁹Pu or ²³⁵U Metal

One postulated scenario of concern is melting of fissile material during the application of the ISV process and the formation of a critical configuration.

Some plutonium might have originally been deposited in metallic form, specifically that associated with metal crucibles (metal waste matrix) and nonmetal molds and crucibles (graphite, glass/slag waste matrices). Any plutonium or uranium disposed of in metallic form is expected to at least have an outer oxide film. Small metal pieces are expected to be completely oxidized (ANS 1980).

Plutonium readily oxidizes in air. Calculations of oxidation rates for 1200 g of ²³⁹Pu metal, as a single sphere surrounded by water, show that after 27 years not all of the metal would be oxidized (see Appendix B6). However, if the metal were divided into many pieces, it would be. In addition, the assumption of a spherical shape with minimum surface area is overly conservative. Flat shapes have more surface area, division of the material into more than one piece increases surface area, and most of the material was in the form of contamination (i.e. finely divided). ISV will not reduce the plutonium back to a metal. The oxide is very stable.

The oxidation potentials for U and Pu are sufficiently high that ISV would not result in reduction of the oxides to the metal. The melting temperature of PuO₂ is 2400°C and the maximum temperatures expected in ISV are about 2000°C. Rather, the lower melting temperature of the ISV melt (1100-2000°C) will disperse the areas of concentrated actinide concentrations, due to the convective currents. The oxide would thus not melt and form a pool. PuO₂ can be reduced in the presence of Ta or Ca to form a slag. However, the process is not very effective. In addition, there is no Ta in the soil at INEEL and Ca is present only as the oxide. There is actually a good amount of CaO in the soil, but the fact that it is already oxidized means it does not seek after the oxygen from PuO₂. Therefore, the driving force for reduction of PuO₂, via Ca, is not in the waste.

The oxide is generally an insoluble form and will be made more insoluble by heating processes such as ISV. As stated previously, some melting of isolated regions of metal could occur if non-oxidized metal pieces exist. However, no credible concentration mechanism for a large amount of fissile material has been identified.

The minimum critical and limiting mass for ²³⁹Pu in a moist-oxide form (1.5 wt% H2O), rather than a solution, is 10.2 kg (LANL 1996) for a system at full density and fully reflected by water. The total quantity of fissile isotopes buried at the SDA has been estimated to be about 350 kg of actinides. Thus, if all the Pu were concentrated in the entire SDA, it would be about 35 times the minimum critical mass for a moist oxide. The fissile material is mostly dispersed at low concentration throughout the waste. Fissile material exists primarily as contamination on the waste material. A few items may potentially contain larger amounts of fissile material, such as filters and graphite material. These materials make up a small percentage of the total waste material both by mass and volume (Clements 1982).

Prior to excavation in the OU 7-10 area, probe holes were put into the SDA in order to gather data relating to the buried waste. As part of this probe hole logging data collection effort, radiation measurements were compiled in an attempt to determine the fissile content in specific locations of this area. From these measurements, a single area believed to contain graphite was estimated to have from 547 g of ²³⁹Pu (probability of unlikely) to 2217 g of ²³⁹Pu (probability of extremely unlikely), based upon conservative calculational approximations (Peatross 2003). Therefore, an area of higher fissile concentration within the SDA cannot be ruled out as impossible. Most likely the material is dispersed in the waste matrices and has undergone oxidation from any metal state.

Therefore, based upon the low overall concentrations of fissile material within the waste, in conjunction with the likelihood that oxidation of any metal has occurred, and the difficulty in reducing oxide to metal form due to ISV, the formation of a critical system (due to the melting and concentration of plutonium metal within the waste matrices of the SDA) is not credible.

6.2 Moderation of Pu by Mixing with Organic Materials such as Polyethylene

The next set of postulated configurations regarding the ISV process, and what physically happens to the waste matrices within the SDA, considers the combination of plutonium and polyethylene. During the ISV process, the temperatures are high enough to drive off organic compounds such as polyethylene, as well as any liquids in the waste melt zone. Most of the waste contained in the melt zone will melt and/or vaporize at their respective melting and vaporization points. Out of the present waste matrices, the most reactive waste form would be polyethylene when combined with plutonium.

In the case of polyethylene, several things must happen in order to cause a criticality during heating of buried waste. Polyethylene first must melt during the initial phase of ISV before temperatures reach levels sufficient to destroy it. The melted polyethylene must selectively entrain or combine in a homogeneous fashion with the fissile isotopes (²³⁹Pu or ²³⁵U). The melted plastic and fissile material must then flow and concentrate in a single area, or from one area to another. This arrangement must be of sufficient concentration and proper shape to moderate neutrons sufficiently to cause a criticality.

Polyethylene, cellulose, and graphite are present in some waste matrices, and represent effective carbon-based neutron moderators and reflectors (Paxton and Pruvost 1986). Polyethylene is superior to water as a neutron reflector/moderator. Polyethylene is a thermoplastic, which melts at 85–110°C. The exact temperature varies with physical properties such as the density, cross-linking frequency, and the degree of crystallinity. Cellulose decomposes at 260-270°C, rather than melt (CRC). Graphite, also an effective moderator, does not melt or decompose, but reacts with oxygen at 110°C or sublimes at 3,652– 3,697°C (CRC). Virtually all moderators (except graphite), including water and most organic materials, leave the heated area undergoing volatilization and destruction (combustion, if oxidizer is present; or pyrolysis, if oxidizer is absent). Polyethylene plastic begins melting with the water vaporization and is completely melted after the water is gone. The moderating water will not be present by the time polyethylene has had sufficient time to melt and pool. Polyethylene is not likely to concentrate fissile material to any extent because it will continue to flow until it pyrolyzes or volatizes. The solubility of fission products in molten polyethylene plastics is likely to be very low, based on the insolubility of most metals, including Pu in aliphatic nonpolar organic materials. Polyethylene, even in larger quantities, does not have the ability to entrain or dissolve appreciable amounts of fissile material, nor does it have any concentrating capacity.

Polyethylene is very viscous during a slow melt. The speed of the heating (ISV might be slow or fast) would determine whether the polyethylene will melt and flow before it is vaporized or pyrolyzed. Polyethylene fluidity in the temperature range between melting and decomposition is low. Although there could be localized movement of molten polyethylene, there will be little if any movement within the waste (a must for postulating the sufficient concentration of fissile material). Moderation from this material is thus not realistically credible for multiple containers on a pit-wide basis. For an optimum polyethylene mixture/solution, the minimum critical mass is less than for water and is estimated to be about 310 g, for a sphere 26 cm in diameter (9.2 L) and ²³⁹Pu concentration of 0.034 g/cc (See Appendix B). This case consisted of PuO₂, combined with polyethylene, with both constituents modeled at the maximum theoretical densities. The sphere was then fully reflected by a 200-cm-thick sphere of vitrified

soil. This set of cases shows the minimum mass that can be made critical and is by no means representative of the expected conditions and configurations within the SDA.

A series of calculational models evaluated a single overloaded 55-gallon drum that contained polyethylene waste material. Moderation of ²³⁹Pu by polyethylene was evaluated here in a series of four single drum problems, taking no credit for the ¹⁰B in the soil. The two configurations modeled 1,200 g of Pu in the form of an oxide. This value was chosen as a bounding condition based upon previous assay data identifying drums with FGE loading in excess of 380 g. The specific waste types were filter media, magnesium oxide heels, grit, tantalum, glass, and molten salt. These drums have since been reassayed and were found to have FGE loading less than 380 g in every case. There are no drums that have been identified by aboveground assaying that contain greater than 380 g FGE. Therefore, 1200 g of Pu, as an oxide in a single drum, is conservative and bounding for the expected configurations to be encountered during the ISV process.

In each problem, the medium outside of the single drum was vitrified soil. In each case, the excess volume in the single drum was filled with full-density polyethylene. The combination of the vitrified soil outside the drum, and the remaining polyethylene in the drum, provided full reflection for the fissile system. Problem Drm05A modeled a single 55-gallon drum containing 65.1% porous PuO₂ (4 g/cc) with polyethylene filling the pores, in the form of a slab at the bottom of the drum. This slab form was chosen to conservatively represent the accumulation of fissile material at the bottom of an intact drum. The accumulation is based upon the density differences between the heavier fissile material and the lighter waste materials. This porosity corresponds to the typical observed density of PuO₂ powder. This was done to address a localized accumulation of fissile material intermixing with polyethylene within a 55-gallon drum (or a localized area in the waste zone), as a result of the application of the ISV process. In reality, the PuO₂ would be mixed with other waste materials. The result was $k_{\rm eff} \pm 1\sigma = 0.3934 \pm 0.0020$. Problem Drm06A used 65.1% porous PuO₂ with polyethylene in the pores and with the polyethylene-impregnated oxide in the shape of a sphere. The result was $k_{\rm eff} \pm 1\sigma = 0.5591 \pm 0.0025$.

Although PuO_2 does not generally dissolve in aliphatic hydrocarbons, it is prudent to examine the consequences of oxide dispersed in polyethylene in a realistic geometry within a buried drum, and determine the concentration for optimum moderation. The mass of Pu was chosen such that at optimum concentration the system would have k_{eff} about or slightly greater than 0.95. The material was assumed to be in a slab located at the bottom of the drum. The remaining volume in the drum was filled with polyethylene at full density. The soil outside was taken as vitrified soil. Initial calculations showed that somewhat less than 700 g of Pu as oxide would be about the limiting amount of Pu in the drum that would be safely sub critical for optimum moderation (see Figure 1). If the mixture were formed into a sphere within the polyethylene, the critical value would be about 310 g (See Appendix B). Figure 1 shows the curve of k_{eff} versus Pu concentration for the slab with 700 g Pu as oxide in polyethylene. The optimum moderation occurs for about 0.02 g Pu/cc where $k_{eff} \pm 1\sigma = 0.9631 \pm 0.0024$. The spherical arrangement was not studied further, as there is no known mechanism for producing it.

These calculations demonstrate the fissile masses necessary to postulate a critical system composed of plutonium and polyethylene, in conjunction with the optimal geometry, reflection conditions, fissile concentration, and lack of diluent/absorber material. The amount of fissile mass necessary in a localized area, and the concurrent conditions necessary, lead to the conclusion that the formation of a critical system due to the initial application of the ISV process is not credible.

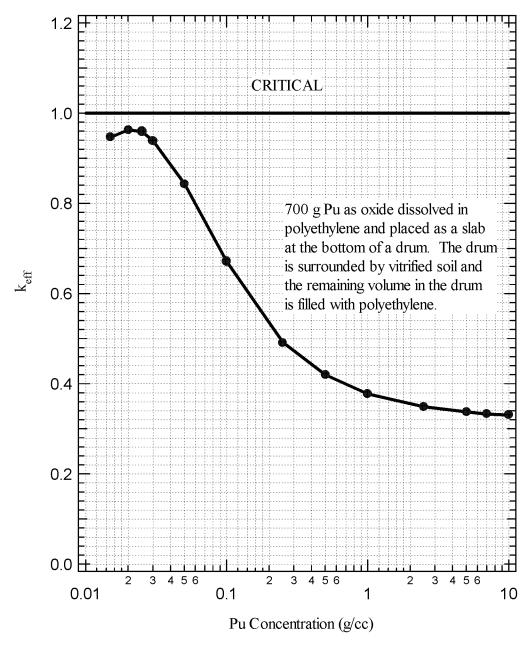


Figure 1. Calculations of $k_{\rm eff}$ for a slab of 239 PuO₂ mixed with polyethylene located at the bottom of a drum surrounded with vitrified soil. The remaining volume of the drum is filled with full density polyethylene.

6.3 Distribution of Plutonium in a Vitrified Matrix

The vitrification process produces a large block of vitrified material. This large vitrified block will replace the current waste zone. The ISV process will set up convective currents within the melt zone that will disperse the fissile material in a homogenous manner throughout the melt zone and eventually the vitrified matrix. The purpose of this section is to address the criticality safety of the vitrified matrix. The following paragraphs describe the various computational models and the respective results from these

models in order to address this issue. Additionally, the presence of graphite within the waste zone and the reactivity effects upon the final vitrified matrix due to its presence is evaluated.

A series of infinite slab problems were thus studied assuming the soil to contain about 70.1 wt% SiO₂ (the actual analysis after ignoring trace materials), 80 wt% SiO₂, 90 wt% SiO₂, or pure silica. Boron in the soil was conservatively assumed to be entirely ¹¹B. The ¹¹B isotope has a much lower thermal absorption cross section than the ¹⁰B isotope. In naturally occurring boron, approximately 19 wt% of the element is composed of ¹⁰B. By modeling the boron present in the soil as completely composed of ¹¹B, the mixture is more reactive from a neutronics standpoint.

The Pu content was taken as oxide mixed in vitrified soil for a range of 0.5 g Pu/l to 2.5 g Pu/l. The vitrified soil was modeled with the same constituents as the dry uncompacted soil, but at a higher density. Dry uncompacted soil has a density modeled as 1.43 g/cc, with vitrified soil having a density modeled as 2.38 g/cc. This scenario represents the homogenizing effect of ISV. The slab thickness was taken as either 10 or 14 ft. The slab was bounded below by a 4-ft-thick layer of clean, vitrified soil and on the top by 6 ft of dry porous soil. The clean, vitrified soil below, and the topsoil above the vitrified block were modeled as not containing any plutonium. Low contamination levels of plutonium might be present in these two media, but in much lower abundance than within the waste matrices. For the cases of 80 and 90 wt% SiO₂, the relative abundance of the other oxides present were unchanged and SiO₂ was added to the mixture. It was assumed that pure SiO₂ was added to the mixture and the material stirred to produce a uniform mixture. Although this would give a continuously changing density, it was found that such density change produced minor effects, so the soil density was not adjusted as the silica was added. Standard handbooks give a maximum density of about 2.32 g/cc for silica. However, for the pure silica case, a density of 2.38 g/cc was used to be consistent with the vitrified soil.

Table 4 and Figure 2 show the results. It can be seen that even for 2.0 g Pu/l with actual soil, the scenario would be sub critical. Even for pure silica and a concentration of 1.0 g Pu/l, the scenario would be sub critical. This latter case would require 634.5 kg of Pu in a $40 \text{ ft} \times 40 \text{ ft}$ area of the pit, which is well beyond a reasonable expected amount. For the SiO_2 content to be as high as 90 wt% would require a 28% error in the soil analysis. Since the soil was mechanically added in the burial process, there is no possibility of a geological anomaly in the mixture.

Table 4. Infinite horizontal slab of vitrified soil of various concentrations of SiO₂ mixed with various concentrations of Pu as oxide.

Prob. Name	Slab Thickness (ft)	Conc (g Pu/l)	Medium	$ m k_{eff}\pm 1\sigma$
U01-01	10	0.5	$70.1 \text{ wt}\% \text{SiO}_2$	0.3347 ± 0.0005
U02-01	10	1.0	70.1 wt% SiO ₂	0.5632 ± 0.0008
U03-01	10	1.5	$70.1 \text{ wt}\% \text{ SiO}_2$	0.7239 ± 0.0010
U04-01	10	2.0	$70.1 \text{ wt}\% \text{ SiO}_2$	0.8379 ± 0.0012
U05-01	10	2.5	$70.1 \text{ wt}\% \text{ SiO}_2$	0.9308 ± 0.0011
U01-02	14	0.5	70.1 wt% SiO ₂	0.3461 ± 0.0005
U02-02	14	1.0	70.1 wt% SiO ₂	0.5836 ± 0.0009
U03-02	14	1.5	$70.1 \text{ wt}\% \text{SiO}_2$	0.7492 ± 0.0010
U04-02	14	2.0	70.1 wt% SiO ₂	0.8720 ± 0.0010
U05-02	14	2.5	70.1 wt% SiO ₂	0.9606 ± 0.0011

Table 2. (continued).

Prob. Name	Slab Thickness (ft)	Conc (g Pu/l)	Medium	$ m k_{eff}\pm 1\sigma$
	(-4)	(8)		
U01-05	10	0.5	80 wt% SiO ₂	0.3858 ± 0.0006
U02-05	10	1.0	80 wt% SiO ₂	0.6369 ± 0.0009
U03-05	10	1.5	$80 \text{ wt}\% \text{SiO}_2$	0.8119 ± 0.0012
U04-05	10	2.0	$80 \text{ wt}\% \text{SiO}_2$	0.9320 ± 0.0013
U05-05	10	2.5	$80 \text{ wt}\% \text{SiO}_2$	1.0209 ± 0.0013
			0 (0) 0	
U01-06	14	0.5	$80 \text{ wt}\% \text{SiO}_2$	0.3995 ± 0.0005
U02-06	14	1.0	$80 \text{ wt}\% \text{SiO}_2$	0.6598 ± 0.0009
U03-06	14	1.5	$80 \text{ wt}\% \text{SiO}_2$	0.8366 ± 0.0011
U04-06	14	2.0	$80 \text{ wt}\% \text{SiO}_2$	0.9620 ± 0.0011
U05-06	14	2.5	80 wt% SiO ₂	1.0526 ± 0.0014
****	1.0	0.7	0.0 10/ 0.0	0.4697.1.0.0006
U01-07	10	0.5	90 wt% SiO ₂	0.4625 ± 0.0006
U02-07	10	1.0	90 wt% SiO ₂	0.7402 ± 0.0010
U03-07	10	1.5	90 wt% SiO_2	0.9244 ± 0.0012
U04-07	10	2.0	90 wt% SiO ₂	1.0421 ± 0.0012
U05-07	10	2.5	90 wt% SiO ₂	1.1283 ± 0.0012
U01-08	14	0.5	90 wt% SiO ₂	0.4798 ± 0.0006
U02-08	14	1.0	90 wt% SiO ₂	0.7665 ± 0.0010
U03-08	14	1.5	90 wt% SiO ₂	0.7663 ± 0.0010 0.9550 ± 0.0012
U04-08	14	2.0	90 wt% SiO ₂ 90 wt% SiO ₂	0.9330 ± 0.0012 1.0796 ± 0.0011
	14	2.5	90 wt% SiO ₂ 90 wt% SiO ₂	
U05-08	14	2.3	90 Wt/6 SIO ₂	1.1695 ± 0.0011
U01-09	10	0.5	100 wt% SiO ₂	0.5892 ± 0.0009
U02-09	10	1.0	$100 \text{ wt}\% \text{ SiO}_2^2$	0.8975 ± 0.0011
U03-09	10	1.5	100 wt% SiO ₂	1.0817 ± 0.0014
U04-09	10	2.0	$100 \text{ wt}\% \text{ SiO}_2$	1.1938 ± 0.0015
U05-09	10	2.5	$100 \text{ wt}\% \text{SiO}_2$	1.2679 ± 0.0015
000 07	10	2.0	100 ((0,00000	1,20,7 = 0,0010
U01-10	14	0.5	100 wt% SiO ₂	0.6142 ± 0.0007
U02-10	14	1.0	100 wt% SiO ₂	0.9320 ± 0.0009
U03-10	14	1.5	100 wt% SiO ₂	1.1178 ± 0.0012
U04-10	14	2.0	100 wt% SiO ₂	1.2339 ± 0.0011
U05-10	14	2.5	100 wt% SiO ₂	1.3132 ± 0.0012

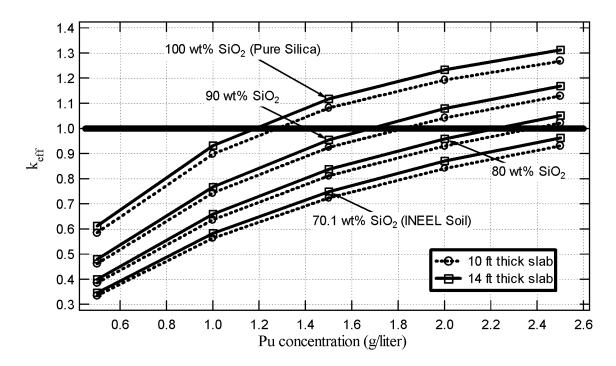


Figure 2. k_{eff} versus Pu concentration for an infinite slab of vitrified soil, mixed with PuO₂ for two values of slab thickness and varied Pu concentrations. The slab is bounded by 4 ft of vitrified clean soil below, and by 6 ft of dry, porous, clean soil above. The four sets of curves are for varied SiO₂ content in the soil.

The case discussed previously evaluated systems with a finite height but infinite in both horizontal directions. Case U05-02 (2.5 g/L Pu in vitrified soil) was also evaluated with finite dimensions in the horizontal directions. These finite systems were reflected by vitrified soil on each of the horizontal faces and the bottom surface with topsoil used as the reflector material on the top surface. The purpose of this set of computational models was to determine the point at which a 14-foot-thick slab becomes infinite in extent in the horizontal directions. Once a finite extent is determined that is equivalent to an infinite extent, an actual mass of plutonium can be calculated. The results from these cases along with a comparison to the result for the infinite by infinite by 14 foot case (U05-02) are given in Table 5.

Table 5. Finite slabs of vitrified soil, 2.5 g/l concentration of Pu as oxide. The slab is bounded by vitrified soil, 4-ft-thick below, and by dry porous soil 6 ft thick above. The slab is 14 ft thick. No credit is taken for ¹⁰B.

Prob. Name	Slab Dimensions (ft)	Mass of Pu Present in Block (kg)	$ m k_{eff}\pm 1\sigma$
U05-02_fin1	10 x 10 x 14	99.1	0.8270 ± 0.0007
U05-02_fin2	15 x 15 x 14	223.0	0.8926 ± 0.0006
U05-02_fin3	$20 \times 20 \times 14$	396.4	0.9198 ± 0.0005
U05-02_fin4	$30 \times 30 \times 14$	892.0	0.9404 ± 0.0005
U05-02_fin5	40 x 40 x 14	1585.7	0.9491 ± 0.0005
U05-02_fin6	50 x 50 x 14	2477.7	0.9533 ± 0.0005
U05-02_fin7	60 x 60 x 14	3567.9	0.9574 ± 0.0005
U05-02	Inf x Inf x 14	NA	0.9606 ± 0.0011

It should be noted that similar results have been obtained for comparable infinite and finite SiO_2 systems in past evaluations.

An additional moderator present in some of the drums is graphite from contaminated molds. It is not known how much is present. Accordingly, a series of infinite slab problems were run using vitrified soil with 10-wt% carbon added. The density change, due to the carbon addition, was ignored. The slab was bounded above by 6 ft of dry, uncompacted soil and below by 4 ft of vitrified soil. Slab thicknesses of 10 ft and 14 ft were evaluated, and the Pu concentration was varied from 0.5 g Pu/l to 2.5 g Pu/l. For the 10-ft-thick slab and the 14-ft-thick slab, the scenarios were safe for concentrations up to 2.0 g Pu/l. Table 6 shows the results. It is highly unlikely that enough graphite would be present in the pit for such scenarios to actually occur, and the amount of Pu required is not credible.

Table 6. Infinite slab of vitrified soil with 10 wt% C added, and with various concentrations of Pu as oxide. The slab is bounded by vitrified soil 4 ft thick below, and by dry porous soil 6 ft thick above. The slab is either 10 ft thick or 14 ft thick. No credit is taken for ¹⁰B.

Prob. Name	Slab Thickness (ft)	Conc (g Pu/l)	$k_{ m eff} \pm 1\sigma$
Q01-01	10	0.5	0.3494 ± 0.0005
Q02-01	10	1.0	0.5908 ± 0.0008
Q03-01	10	1.5	0.7645 ± 0.0009
Q04-01	10	2.0	0.8889 ± 0.0009
Q05-01	10	2.5	0.9856 ± 0.0013
Q01-02	14	0.5	0.3601 ± 0.0005
Q02-02	14	1.0	0.6086 ± 0.0007
Q03-02	14	1.5	0.7848 ± 0.0010
Q04-02	14	2.0	0.9112 ± 0.0010
Q05-02	14	2.5	1.0132 ± 0.0012

6.4 Comparison between PuO₂ distributed in Vitrified Soil or Water Saturated Soil

The previous studies in this evaluation analyzed plutonium at various concentrations within a large homogeneous block of vitrified material. A set of calculational models was developed to compare the reactivity effects between PuO₂ dispersed in vitrified soil or water-saturated soil. These models evaluated higher localized fissile masses in optimum spherical geometric configurations. The purpose of this set of calculations is to show the decrease in reactivity between like systems, due to the application of the ISV process, as opposed to no treatment and the possibility of future water in-leakage into the waste zone.

Studies were made using the MCNP code to calculate $k_{\rm eff}$ for a range of ²³⁹Pu concentrations in a mixture contained in a sphere and fully reflected. Initial studies were made for single spheres containing a mixture of either ²³⁹PuO₂ and vitrified soil, or ²³⁹PuO₂ and water-saturated nominal soil (SDA soil has a porosity of 40%). Curves were calculated for spheres with a mass of 1200 g Pu mixed with either vitrified soil or water-saturated soil. In all of these cases, the medium surrounding the spheres was 5 ft of 40% porous, water-saturated soil. These results are given in Table 7 and shown graphically in Figure 3.

Table 7. MCNP calculations of k_{eff} versus Pu concentration for 1200 g Pu as oxide and mixed with either vitrified soil or 40% porous saturated soil, surrounded by saturated soil. No 10 B present.

	Conc (g/cc)	Pu & Vitr. Soil		Pu & Sat. Soil	
	²³⁹ Pu	Prob. Name	$k_{\rm eff}\!\pm1\sigma$	Prob. Name	$k_{\mathrm{eff}} \pm 1\sigma$
(0.015	Y0101A	0.3521 ± 0.0017	Z0101A	0.9269 ± 0.0023
(0.020	Y0201A	0.3616 ± 0.0017	Z0201A	0.9269 ± 0.0028
(0.025	Y0301A	0.3620 ± 0.0019	Z0301A	0.9230 ± 0.0023
(0.030	Y0401A	0.3632 ± 0.0017	Z0401A	0.9115 ± 0.0028
(0.050	Y0501A	0.3641 ± 0.0022	Z0501A	0.8488 ± 0.0031
(0.100	Y0701A	0.3522 ± 0.0020	Z0701A	0.7389 ± 0.0026
(0.250	Y0801A	0.3254 ± 0.0019	Z0801A	0.5829 ± 0.0025
(0.500	Y0901A	0.3093 ± 0.0020	Z0901A	0.4878 ± 0.0026
1	1.000	Y1101A	0.3038 ± 0.0015	Z1101A	0.4220 ± 0.0022
2	2.500	Y1201A	0.3281 ± 0.0019	Z1201A	0.3799 ± 0.0020
5	5.000	Y1501A	0.3834 ± 0.0016	Z1501A	0.4031 ± 0.0019
7	7.000	Y1701A	0.4255 ± 0.0017	Z1701A	0.4335 ± 0.0017
10	0.00	Y2001A	0.4837 ± 0.0017	Z2001A	0.4869 ± 0.0018

As shown in the graph, the 1200 g curves merge at 10 g Pu/cc since that is almost pure oxide with no vitrified or saturated soil left in the mixture. At the high-density end of the curves, the density of the Pu is the dominant factor. As moderator is introduced, the reactivity increases until a peak in k_{eff} occurs. For further dilution of the mixtures, absorption becomes the dominant factor and k_{eff} then decreases, however there is no credible mechanism for preferentially concentrating the PuO₂ mixed with water-saturated soil into the required spheres and surrounding them with water-saturated soil as a medium. It is not credible for a criticality to occur in the SDA due to water intrusion for waste in its current configuration. Activities such as ISV, which would remove plastic material, would also eliminate the water that is necessary to formulate a critical system.

6.5 Arrays of Drums Containing Pu distributed in Vitrified Soil and Saturated Soil

The previous cases analyzed vitrified blocks and compared vitrified configurations to similar water-saturated soil configurations. The premise of the ISV process is that a uniform melt occurs in the waste zone as it grows outwardly. The purpose of this set of cases is to evaluate the reactivity associated with postulated localized vitrification in overloaded drums based upon past retrieval history. It is expected that most buried drums lack integrity. These cases assume the drums are intact to provide a basis to evaluate ordered arrays of vitrified localized pockets of fissile material. This will address any concerns associated with localized vitrification and the failure of the convective currents to adequately disperse the fissile material into the melt matrix.

PuO₂ Distibuted in Vitrified vs. Saturated Soil

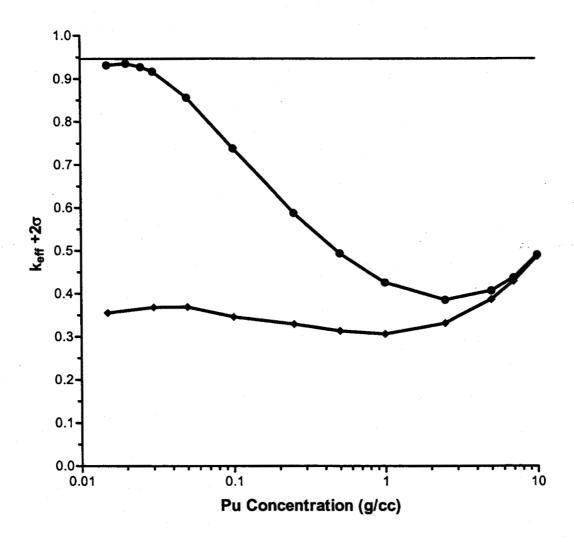


Figure 3. Calculations of k_{eff} for spheres of 239 PuO₂ mixed with either vitrified soil or 40% porous water-saturated soil, and reflected by 40% porous water-saturated soil.

For this study, a square array of 11 drums by 11 drums stacked three drums high was modeled. The drums were modeled as cuboids in which the height of the drum was preserved, with the horizontal dimensions of the modeled cuboid equivalent to the original diameter of the drum. This cuboid arrangement allowed for a densely packed array of "drums" to be evaluated. Initial drum disposal operations called for the placement of drums into the trenches in a neat orderly array. Due to ALARA concerns, the drums were eventually dumped randomly into the pit. In the cases where drums were stacked, degradation of the drum material, due to the burial timeframes, would lead to arrays of drums similar to those expected when the drums were just dumped into the burial pits. A conservative configuration, such as a stacked regular array, gives a tighter packing fraction than the more expected actual configuration. The area of the model is approximately 20 ft × 20 ft. For the application to actual waste material, the area may be about 30 ft in diameter.

The materials in each drum were treated as a single sphere surrounded by soil. For most of the drums, a loading of 200 g Pu as oxide (the nominal limit) mixed with vitrified soil (70.1% SiO₂ mixture) was assumed. In the center of the array in the central plane, four overloaded drums were located adjacent to each other. One drum contained 1200g Pu as oxide mixed with vitrified soil (70.1% SiO₂ mixture) in a single sphere. The other three contained 300 g Pu as oxide with vitrified soil (70.1% SiO₂ mixture) in single spheres. These fissile amounts adequately bound the expected fissile loading within the buried waste drums in the SDA. All four spheres were clustered in contact with each other without any constraints that would be imposed by the geometry of the drums. The surrounding medium was either water-saturated soil or vitrified soil. The concentration of the Pu was varied from 0.015 g Pu/cc to 10 g Pu/cc. Table 8 shows the results. In addition, an array was evaluated with Pu as oxide mixed with saturated soil and the same medium. Table 9 shows the results. The purpose of these calculations was to show the relative reactive k_{eff}'s achieved when combining the PuO₂ with saturated soil compared to PuO₂ in vitrified soil. The assumptions in these models are very conservative and are performed to show that even for such unrealistic assumptions, a critical system is not formed.

Figure 4 shows that the curve with PuO_2 mixed with water-saturated soil in a medium of water-saturated soil is the most reactive and shows the characteristic shape. At high concentrations, the density of the Pu is the dominant factor. As moderator is introduced, the reactivity increases until a peak in $k_{\rm eff}$ occurs. For further dilution of the mixtures, absorption becomes the dominant factor and $k_{\rm eff}$ then decreases. Except for high-density cases, 239 PuO₂ mixed with water-saturated soil is more reactive.

All of these are not credible situations. The ISV process will remove and exclude water from the vitrified medium. Therefore, the PuO_2 and water-saturated soil models will not exist in the final configuration. There is no credible mechanism to concentrate and mix enough PuO_2 with water-saturated soil into the required spheres, thus creating a critical system.

Table 8. $11 \times 11 \times 3$ array of drums, fissile material mixed in vitrified matrix, nominal loading per drum 200 g Pu. Four adjacent drums near the center of the array are abnormal (1200 g Pu as oxide mixed with vitrified soil, and the other three contain 300 g Pu as oxide mixed with vitrified soil). The abnormal spheres are clustered together. The surrounding medium is either vitrified soil or 40% porous, water-saturated soil.

Conc	Prob.	Vitr. Soil Medium Reflector	Prob.	Sat. Soil Medium Reflector
(g/cc)	Name	$k_{ m eff} \pm 1\sigma$	Name	$k_{ m eff} \pm 1\sigma$
0.015	X0106A	0.2904 ± 0.0009	X0101A	0.4297 ± 0.0012
0.030	X0406A	0.2476 ± 0.0010	X0401A	0.4589 ± 0.0012
0.050	X0506A	0.2254 ± 0.0009	X0501A	0.4718 ± 0.0011
0.100	X0706A	0.2013 ± 0.0009	X0701A	0.4621 ± 0.0013
0.250	X0806A	0.1862 ± 0.0008	X0801A	0.4307 ± 0.0011
0.500	X0906A	0.1924 ± 0.0008	X0901A	0.4082 ± 0.0012
1.000	X1106A	0.2156 ± 0.0007	X1101A	0.3949 ± 0.0011
2.500	X1206A	0.2810 ± 0.0007	X1201A	0.4020 ± 0.0012
5.000	X1506A	0.3678 ± 0.0007	X1501A	0.4504 ± 0.0011
7.000	X1706A	0.4225 ± 0.0008	X1701A	0.4889 ± 0.0011
10.00	X2006A	0.4984 ± 0.0008	X2001A	0.5455 ± 0.0010

Table 9. $11 \times 11 \times 3$ array of drums fissile material mixed in saturated soil, nominal loading per drum 200 g Pu. Four adjacent drums near the center of the array are abnormal (1200 g Pu as oxide mixed with saturated soil, and the other three contain 300 g Pu as oxide mixed with saturated soil). The abnormal spheres are clustered together. The surrounding medium is 40% porous, water-saturated soil.

Conc (g/cc)	Prob. Name	$ m k_{eff}$	
0.015	W 0101 A	0.9384 ± 0.0014	
0.020	W0201A	0.9520 ± 0.0015	
0.025	W0301A	0.9483 ± 0.0017	
0.030	W0401A	0.9440 ± 0.0018	
0.050	W0501A	0.9008 ± 0.0018	
0.100	W0701A	0.8124 ± 0.0016	
0.250	W0801A	0.6855 ± 0.0016	
0.500	W0901A	0.5971 ± 0.0017	
1.000	W1101A	0.5227 ± 0.0012	
2.500	W1201A	0.4697 ± 0.0012	
5.000	W1501A	0.4772 ± 0.0012	
7.000	W1701A	0.4991 ± 0.0011	
10.00	W2001A	0.5465 ± 0.0011	

These models show that even with these conservative assumptions, a configuration of localized high fissile areas in a vitrified media will not create a critical system and will be less reactive than a system that is made up of water-saturated soil and fissile material.

6.6 Melt Expulsion

The possibility of a melt expulsion and any criticality implications need to be addressed as part of this overall evaluation of the ISV process. A melt expulsion is a possible upset condition from the heating of a volatile chemical or creation of a steam pocket in the waste zone, resulting in an expulsion of material out of the waste area. This issue, and its relation to criticality safety, will be addressed here.

Melt expulsion would scatter any entrained fissile material. This would reduce reactivity. Dynamic disruption is proposed as a possible method to prevent pressure buildup in sealed drums. Steel rods are driven through the waste seam at intervals to puncture buried containers. This, however, will not deal with buried high-pressure gas cylinders. The safety concern in melt expulsion is the scattering of radioactive material and damage to equipment rather than criticality. There is no criticality concern with melt expulsion.

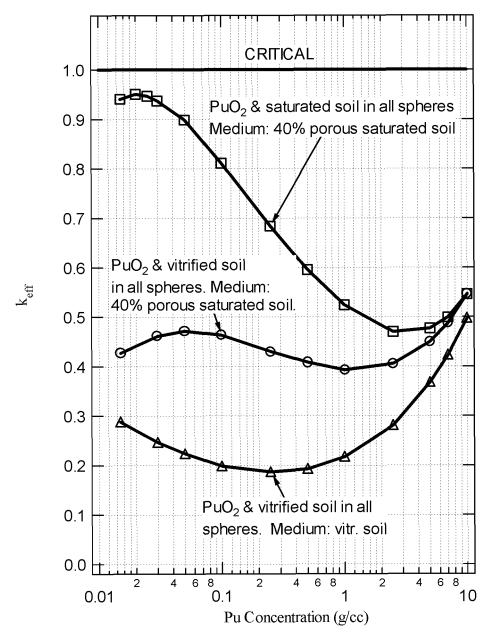


Figure 4. Graphical Representation of Results from Tables 8 and 9.

6.7 Flooding and/or Water Re-entry

The concern of flooding or water reentry into the SDA, following the ISV application, will be discussed here. The issue that needs to be addressed is whether or not the application of the ISV process increases the probability for the formation of a critical system within the vitrified matrix, as opposed to water intrusion into the current waste configuration. The answer to this is no, with the justification provided below.

In 1969, local runoff from rapid spring thaws caused flooding that covered part of the SDA with water. A 4.6 m (15 ft) dike has since been constructed around the SDA to prevent future flooding. In the case of ISV, there would not be sufficient void volume in the final vitrified material for intrusion of extra

water, such that it would be intimately mixed with the entrained fissile material. As previously discussed, the ISV process will form a melt that will disperse the fissile material as opposed to concentrating it. Therefore, even if some water were to enter the vitrified matrix, it would be still be less reactive than if it were to enter the waste zone prior to the application of ISV. Therefore, the formation of a critical system, due to flooding, is not possible after the application of the ISV process.

6.8 Collection of Fissile Materials in the Off-Gas System

The last aspect of the proposed ISV process that needs to be addressed from a criticality safety standpoint is the possibility to formulate a critical configuration in the off-gas collection system. This will be discussed next.

The off-gas collection system consists of three parts: 1) the off gas hood, 2) a HEPA pre-filter system, and 3) the off-gas treatment system.

The off-gas treatment hood contains the gaseous effluents from ISV processing, provides a confined area for oxidation of the effluents, and directs the effluents to the off-gas system. An off-gas HEPA prefilter system is employed between the off-gas hood and the off-gas treatment trailer. This system consists of propane-fueled gas flame preheater units, high-temperature HEPA filter units, and a bypass line. The housings for the preheater units and the HEPA filter units are bolted into the off-gas line using standard flanges.

The primary components of the off-gas treatment system include a gas cooler, dual wet scrubber systems (with tandem nozzle scrubbers and quenchers), heat exchangers, process scrub tanks, scrub solution pumps, a condenser, three mist eliminators (vane separators), a heater, a dual stage HEPA filter assembly, and a blower system.

Accumulation of sufficient fissile material in the off-gas system to cause a criticality event is not credible. The subsurface nature of planar-ISV is such that most of the fissile material is contained and incorporated in the melt zone itself (approximately 99.9% in the melt, with a large fraction filtered by the overburden) (Farnsworth et al. 1999). The amount of plutonium that migrates from the melt and through the overburden will not be significant. Therefore, a criticality accident is not credible in the off-gas collection system.

7. DESIGN FEATURES AND ADMINISTRATIVE CONTROLS & LIMITS

There are no design features or administrative controls necessary in the application of the ISV process.

8. SUMMARY AND CONCLUSIONS

Various configurations were evaluated to determine if any criticality concerns arise in conjunction with treating the buried waste contained in the SDA with the ISV process. This evaluation consisted of three phases. The first phase was to consider criticality scenarios during the initial application of the ISV process. The second phase consisted of evaluating the final configurations as a result of the application of the ISV process. The third phase was to address ancillary issues relating to ISV and criticality safety. Each of these phases is further described in the remainder of this section.

The first phase consisted of the evaluation of criticality concerns and postulated scenarios during the initial application of the ISV process. They include the formation of a critical system, due to the accumulation of fissile material in the form of a molten metal, and the combination of fissile material and melting polyethylene.

The formation of a critical system, due to the accumulation of fissile material in the form of molten metal, was evaluated. As discussed, in order to form a critical system from the melting of molten metal, it would be necessary to have a large quantity of fissile material in metal form in a relatively localized area within the waste zone. This is not expected. Calculations were made to show that a spherical piece of plutonium metal (on the order of 6 kg) would almost completely oxidize in the amount of time that the waste has been buried at the SDA. If smaller pieces and fragments existed, which is expected in the waste, they would most likely be completely oxidized. The temperature in the ISV process is not sufficient to reduce any fissile material in the form of oxide to metal. Therefore, this scenario was deemed not to be credible.

The formation of a critical system, due to the initial melting of polyethylene and the subsequent mixing with enough plutonium in the geometrical form, concentration, and reflective conditions required, was deemed to be not credible. In order to achieve such a state, the plutonium must somehow capture the plutonium oxide in a very ordered manner as it is beginning to melt and flow. As temperatures increase within the melt zone, the polyethylene (along with other plastic-type materials) will be destroyed and removed from the system. It was shown that an idealized optimum configuration of plutonium and polyethylene could be made critical with as little as 310 g in a spherical configuration and 700 g in a cylindrical configuration conforming to the confines of a drum. The idealized configurations are not realistic with the SDA waste zone. The amount of fissile material, the concentration of fissile material, the geometrical configuration, the reflective conditions, and the lack of diluent/neutronic absorber present in these idealized cases do not by any means represent the configurations within the waste zone. Allowing for the introduction of some realism, modeling of the system as a flattened cylinder increases the necessary fissile mass required to form a critical system above that which is expected in any localized areas of the SDA. Therefore this scenario is not credible.

The second phase consisted of evaluating the final expected waste configuration, due to the application of the ISV process.

As the ISV process is applied, the melt zone grows outward until eventually the desired area has been melted. The probes are then removed and the cooling process begins. The probes are then moved

such that the next melt zone overlaps the previous and in the end creating an eventual large vitrified mass. The melt zone sets up convective currents that tend to disperse the fissile material in a homogenous fashion within the melt region. The final configurations were evaluated through a series of computational models.

The first models consisted of vitrified matrices with finite heights that were infinite in the horizontal directions. The infinite slab problems simulated the final configuration of ISV with a range of SiO_2 content in a vitrified soil matrix. The results for these models showed that concentrations up to 2.0 g Pu/l were safe when the actual soil containing 70.1 wt% SiO_2 is used, and concentrations up to 1.0 g Pu/cc were safe for a vitrified medium of 100 wt% SiO_2 . In order to determine some bounding fissile masses finite models were evaluated. These cases consisted of the 70.1 wt% SiO_2 vitrified soil at a height of 14 ft with an increasing horizontal cross-sectional area from 10X10 ft to 40X40 ft. A Pu concentration of 2.5 g/l was used in these models. It was found that a 30X30X14-ft-block of vitrified material, containing 892 kg of Pu, was safe with a $k_{\rm eff} + 2\sigma = 0.942$. This fissile amount equates to the expected entire plutonium inventory for the SDA in the 20X20X14-ft-block.

In order to address the possibility of localized vitrification, finite arrays of drums were evaluated. In these models the fissile material was concentrated into spherical form in four adjacent drums located physically in contact with each other. The four drums located in close proximity were overloaded with 1200 g of Pu in a single drum, 300 g of Pu in the next three closest drums, and 200 g of Pu in the remaining drums in the array. This model was intended to address the questions associated with an array of ordered drums and the effect ISV would have on the contents. The first consideration is that ISV would remove the drums and cause enough mixing to prevent the hypothetical spheres from forming. Additionally, these are not realistic scenarios since it is known that buried waste that was placed into the SDA in an orderly fashion is no longer orderly. The ISV process would set up convection currents in the melt zone that would create a dispersion of the Pu into the eventual vitrified matrix along with the many other neutron diluents and absorbers present in the buried waste. There is no mechanism related to ISV to postulate the formulation of these geometrically reactive scenarios. However, these conservative models show that the application of the ISV process would not form a critical system in an ordered array of drums containing vitrified material.

Along these lines, similar models were compared that consisted of fissile material dispersed in water-saturated soil. These models show that the vitrified matrix is less reactive than similar configurations of saturated soil.

Other mechanisms were evaluated qualitatively such as melt expulsion, water reentry into the vitrified matrix, and collection of fissile material in the off-gas system. The conclusion from these assessments was that none of these scenarios lend themselves to the credible formation of a critical system.

The conclusion of the evaluation is that there is no credible scenario associated with the ISV process to formulate a critical system.

9. REFERENCES

- ANS 1980, "Plutonium Handbook: A Guide to the Technology", Vol. 1, 1980, Oz Wick, Ed., American Nuclear Society (ANS), La Grange Park, Illinois
- Buelt, J. L., Timmerman, C. L., Oma, K. H., Fitzpatrick, V. F. and Carter, J. G., "In Situ Vitrification of Transuranic Wastes: An Updated Systems Evaluation and Applications Assessment", PNL-4800, Suppl. 1, March 1987, Pacific Northwest Laboratory, Richland, WA 99352
- Callow, R. A., L. E. Thompson, J. R. Weidner, C. A. Loehr, B. P. McGrail, and S. O. Bates, 1991, *In Situ Vitrification Application to Buried Waste Final Report of Intermediate Field Tests at Idaho National Engineering Laboratory*, EGG WTD-9807, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.
- Cember H., 1985 "Introduction to Health Physics", Chapter 12 Criticality. pp 364-383
- Clements, T. L., Jr., 1982 "Content Code Assessments for INEL Contact-Handled Stored Transuranic Wastes", WM-F1-82-021-C3. October, 1982, EG&G Idaho, Inc.
- CRC, 1982"CRC Handbook of Chemistry and Physics", 62nd Ed., Chemical Rubber Company
- Farnsworth, R. K., Henrikson, D. J., Hyde, R. A., Jorgensen, D. K., Mcdonald, J. K., Nickelson, D. F., Pfeifer, M. C., Sloan, P. A., Weidner, J. R., 1999 "Operable Unit 7-13/14 In Situ Vitrification Treatability Study Work Plan", DOE/ID-10667, Idaho National Engineering and Environmental Laboratory.
- INEEL, 1998, *Program Requirements Document for Criticality Safety*, PRD-112, Category 2, Rev. 1, Idaho National Engineering and Environmental Laboratory, Lockheed Martin Idaho Technologies Company (LMITCO), LLC, Idaho Falls, Idaho.
- LANL, 1996, Nuclear Criticality Safety Guide, LA-12808, Los Alamos National Laboratory, (LANL), Los Alamos, New Mexico.
- Nielsen, J.W. 2003, "Validation of Calculations Containing HEW/Graphite and Pu/Polystyrene," INEEL/INT-02-01248, Idaho National Engineering and Environmental Laboratory, March 2003
- Paxton, H. C., Pruvost, N. L., 1986 "Critical Dimensions of Systems Containing ²³⁵U, ²³⁹Pu and ²³³U", 1986 Revision, LA-10860-MS, Los Alamos National Laboratory, July 1987.
- Peatross, R. G., 2003, Radioactive Waste Management complex Safety Analysis Report Addendum J, The OU 7-10 Glovebox Excavator Method Project, INEL-94-0226, SAR-4, Addendum J, Idaho National Engineering and Environmental Laboratory.
- RSIC, 1997, "MCNP4-A General Monte Carlo N-Particle Transport Code, Version 4B," CCC-660/MCNP4B2, Radiation Shielding Information Center (contributed by Los Alamos National Laboratory), Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Tsiboulia A., I. Matveenko, M. Nikolaev, and M. Semenov, 2000, *Critical Experiments with Heterogeneous Compositions of Highly Enriched Uranium, Silicon Dioxide, and Polyethylene*, HEU-MET-MIXED-005, International Handbook of Evaluated Criticality Safety Benchmark Experiments.

Appendix A Materials and Compositions

Appendix A

Materials and Compositions

Table A-1. INEEL soil sample analysis (average of two analyses).

Table A-2. Renormalized INEEL Soil Sample for Dry Material.

110 41141 3000).			
Oxide	Composition (wt%)	Oxide	Composition (wt%)
SiO ₂	62.60	SiO_2	69.936
Al_2O_3	11.85	Al_2O_3	13.239
Fe_2O_3	4.25	Fe_2O_3	4.748
CaO	3.68	CaO	4.111
K_2O	2.99	K_2O	3.340
MgO	1.72	MgO	1.922
Na_2O	1.37	Na_2O	1.531
TiO_2	0.68	TiO_2	0.760
\mathbf{MnO}_2	0.10	\mathbf{MnO}_2	0.112
BaO	0.09	BaO	0.101
ZrO_2	0.05	\mathbf{ZrO}_2	0.056
B_2O_3	0.05	$\mathbf{B}_2\mathbf{O}_3$	0.056
NiO	0.04	NiO	0.044
SrO	0.02	SrO	0.022
Cr_2O_3	0.02	Cr_2O_3	0.022
Total Oxide	89.5%	Total Oxide	100.0%
Moisture	7.5%		

Appendix B Supporting Calculations and Methods

Appendix B

Supporting Calculations and Methods

B-1. CONTAMINATED VITRIFIED SOIL CALCULATIONS

Consider a sample of vitrified soil with volume v_{slag} and density ρ_{slag} . It has mass m_{slag} and is composed entirely of oxides. We know the mass fraction f_i of each oxide in the soil, so we know the mass of each oxide in the sample. Consider SiO₂.

$$m_{SiO_2} = f_{SiO_2} m_{slag}$$
, $M_{SiO_2} = M_{Si} + 2M_o$ and $m_{slag} = \rho_{slag} V_{slag}$ so that

$$m_{Si} = \left(\frac{M_{Si}}{M_{SiO_2}}\right) m_{SiO_2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}}\right) \rho_{slag} V_{slag}$$

Now consider adding PuO₂ with density ρ_{PuO_2} to the soil. Let $\mu = \frac{m_{PuO_2}}{V_{mix}}$ be the concentration (g/cc) of PuO₂ in the mix. Now $V_{mix} = V_{slag} + V_{PuO_2}$ and $m_{PuO_2} = \mu V_{mix}$.

$$m_{mix} = m_{slag} + m_{PuO_2} = m_{slag} + \mu V_{mix} = \rho_{slag} V_{slag} + \mu V_{mix}$$

$$\rho_{mix} = \frac{m_{mix}}{V_{mix}} = \frac{\rho_{slag}V_{slag}}{V_{mix}} + \mu, \quad \rho_{PuO_2}V_{PuO_2} = m_{PuO_2} = \mu V_{mix} \quad \text{and} \quad V_{mix} = \frac{m_{PuO_2}}{\mu}$$

$$V_{mix} = \frac{m_{PuO_2}}{\mu} = V_{slag} + V_{PuO_2} = \frac{\rho_{PuO_2} V_{PuO_2}}{\mu} \quad \text{so that} \quad V_{PuO_2} \left[\frac{\rho_{PuO_2}}{\mu} - 1 \right] = V_{slag}$$
 (1)

Combining (1) with $V_{mix} = V_{slag} + V_{PuO_2}$ gives

$$V_{mix} = V_{slag} \left[\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu} \right] \tag{2}$$

Then

$$\rho_{mix} = \frac{\rho_{slag} V_{slag}}{V_{mix}} + \mu \quad \text{and} \quad \frac{V_{slag}}{V_{mix}} = \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}}\right)$$

so that

$$\rho_{mix} = \rho_{slag} \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right) + \mu \tag{3}$$

From (1) we have
$$V_{PuO_2} = \left(\frac{\mu}{\rho_{PuO_2} - \mu}\right) V_{slag}$$

Let $\xi = \frac{m_{Pu}}{V_{mix}}$ be the concentration (g/cc) of Pu in the mix. We have $m_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) m_{PuO_2}$ so

that
$$\xi = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \frac{m_{PuO_2}}{V_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \mu$$
.

We thus have

$$\mu = \left(\frac{M_{PuO_2}}{M_{Pu}}\right)\xi\tag{4}$$

Now $m_{Si} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \rho_{slag} V_{slag}$ and $m_{mix} = \rho_{mix} V_{mix} = \rho_{mix} V_{slag} \left(\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu} \right)$ so that

$$\frac{m_{si}}{m_{mix}} = \frac{f_{SiO_2}\left(\frac{M_{Si}}{M_{SiO_2}}\right)\rho_{slag}V_{slag}}{\rho_{mix}V_{slag}\left(\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu}\right)} = f_{SiO_2}\left(\frac{M_{Si}}{M_{SiO_2}}\right)\left(\frac{\rho_{slag}}{\rho_{mix}}\right)\left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}}\right)$$

and the weight fraction of Silicon in the mix is

$$F_{\mathrm{Si}} = f_{\mathrm{SiO_2}} \left(\frac{M_{\mathrm{Si}}}{M_{\mathrm{SiO_2}}} \right) \left(\frac{\rho_{\mathrm{slag}}}{\rho_{\mathrm{mix}}} \right) \left(\frac{\rho_{\mathrm{PuO_2}} - \mu}{\rho_{\mathrm{PuO_2}}} \right)$$

$$F_{Pu} = \frac{m_{Pu}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \frac{m_{PuO_2}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \frac{\mu}{\rho_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \left(\frac{M_{PuO_2}}{M_{Pu}}\right) \frac{\xi}{\rho_{mix}} = \frac{\xi}{\rho_{mix}}.$$
 (5)

Now consider the mass fraction of oxygen contributed by the PuO2.

$$m_{PuO_2} = m_{Pu} + m_O^{PuO_2}$$
 so that $\frac{m_{PuO_2}}{m_{mix}} = \frac{m_{Pu}}{m_{mix}} + \frac{m_O^{PuO_2}}{m_{mix}} = F_{Pu} + F_O^{PuO_2}$

Now
$$\mu = \frac{m_{PuO_2}}{V_{mix}}$$
 so that $\frac{m_{PuO_2}}{m_{mix}} = \frac{\mu V_{mix}}{m_{mix}} = \frac{\mu}{\rho_{mix}}$, $\frac{\mu}{\rho_{mix}} = F_{Pu} + F_O^{PuO_2}$ and $F_O^{PuO_2} = \frac{\mu}{\rho_{mix}} - F_{Pu}$

$$\mu = \left(\frac{M_{PuO_2}}{M_{Pu}}\right)\xi \qquad F_O^{PuO_2} = \frac{1}{\rho_{mix}} \left(\frac{M_{PuO_2}}{M_{Pu}}\right)\xi - F_{Pu}$$
but so that we have

Consider the oxygen contribution from SiO₂. We have 2 oxygen molecules/molecule so that the mass fraction in the mix is given by

$$F_{\scriptscriptstyle O}^{\scriptscriptstyle SiO_2} = f_{\scriptscriptstyle SiO_2} \Biggl(\frac{2M_{\scriptscriptstyle O}}{M_{\scriptscriptstyle SiO_2}}\Biggr) \Biggl(\frac{\rho_{\scriptscriptstyle slag}}{\rho_{\scriptscriptstyle mix}}\Biggr) \Biggl(\frac{\rho_{\scriptscriptstyle PuO_2} - \mu}{\rho_{\scriptscriptstyle PuO_2}}\Biggr) = f_{\scriptscriptstyle SiO_2} \Biggl(\frac{2M_{\scriptscriptstyle O}}{M_{\scriptscriptstyle SiO_2}}\Biggr) \phi$$

The oxygen contributions from the other oxides are calculated similarly.

Now consider atomic fractions and total atom density. We have

$$N_{Si} = \frac{m_{Si}(g)}{M_{Si}(g/mole)} N_A(@/mole) \qquad n_{Si} = \frac{N_{Si}}{V_{mix}} = \frac{m_{Si}}{V_{mix}} \frac{N_A}{M_{Si}}$$

but
$$m_{Si} = F_{Si} m_{mix}$$
 so that $\frac{m_{Si}}{V_{mix}} = F_{Si} \frac{m_{mix}}{V_{mix}} = F_{Si} \rho_{mix}$ and thus, in general,

$$n_{Si} = F_{Si} \rho_{mix} \frac{N_A}{M_{Si}}$$

$$n_{cation_i} = F_{cation_i} \rho_{mix} \left(\frac{N_A}{M_{cation_i}} \right)$$
 and $n_{Pu} = F_{Pu} \rho_{mix} \left(\frac{N_A}{M_{Pu}} \right)$ $n_O = \sum_i n_{O_i}$

$$n_{mix} = \sum_{i} n_{cation_i} + n_{Pu} + \sum_{i} n_{O_i}$$

Let β_i be the atomic fraction for cation_i.

$$\beta_{cation_i} = \frac{n_{cation_i}}{n_{mix}}$$
 $\beta_{Pu} = \frac{n_{Pu}}{n_{mix}}$ $\beta_O = \frac{n_O}{n_{mix}}$

Now $m_{SiO_2}^O \equiv$ mass of oxygen from SiO2, and

$$m_{SiO_{2}}^{O} = \left(\frac{2M_{O}}{M_{SiO_{2}}}\right) m_{SiO_{2}} = \left(\frac{2M_{O}}{M_{SiO_{2}}}\right) f_{SiO_{2}} m_{slag} = \left(\frac{2M_{O}}{M_{SiO_{2}}}\right) f_{SiO_{2}} \rho_{slag} V_{slag}$$

$$\frac{m_{SiO_2}^O}{m_{mix}} = \frac{m_{SiO_2}^O}{\rho_{mix}V_{mix}} = \left(\frac{2M_O}{M_{SiO_2}}\right) f_{SiO_2} \frac{\rho_{slag}V_{slag}}{\rho_{mix}V_{mix}} \qquad \frac{V_{slag}}{V_{mix}} = \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}}\right)$$
so tha

$$\frac{\textit{\textit{m}}_{\textit{SiO}_2}^{\textit{O}}}{\textit{\textit{\textit{m}}}_{\textit{mix}}} = \left(\frac{2M_{\textit{O}}}{M_{\textit{SiO}_2}}\right) f_{\textit{SiO}_2} \frac{\rho_{\textit{slag}}}{\rho_{\textit{mix}}} \left(\frac{\rho_{\textit{PuO}_2} - \mu}{\rho_{\textit{PuO}_2}}\right)$$

Calculate the corresponding quantity for the other oxides and PuO_2 and add them up to get F_o .

Procedure

- Specify ξ
- Calculate μ . 2.
- For given $\,^{\rho_{\textit{slag}}}\,,\,^{\rho_{\textit{PuO}_2}}\,,\,$ calculate $\,^{\rho_{\textit{mix}}}\,.$

$$\varphi \equiv \left(\frac{\rho_{slag}}{\rho_{mix}}\right) \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}}\right)$$
4 Calculate

- 4.
- Given the mass fractions of the oxides in the soil, calculate the mass fractions F_{cation_i} of the cations 5. in the mix.
- Calculate the mass fraction for the plutonium: F_{Pu} . 6.
- Calculate the atomic densities n_{cation_i} from the mass fractions. 7.
- Calculate the total atom density n_O for oxygen by summing the atomic densities n_i^O from each 8. oxide.
- 9. Calculate the total atom density for the mixture by summing the atom densities of the constituents.
- Calculate the atomic fractions $\beta_{\textit{cation}_i}$, $\beta_{\textit{Pu}}$ and $\beta_{\textit{O}}$. 10.

B-2. SATURATED POROUS SOIL CALCULATIONS

Consider V_S of solids with mass $m_s = \rho_s V_s$ and with 40% porosity. We have $V_s = 0.6 V_T$ where V_T is the total volume occupied by the porous soil. We then have

$$V_T = \frac{V_s}{0.6} = \frac{5}{3}V_s$$

The water filling the pores occupies the rest:

$$V_w = 0.4V_T = 0.4 \left(\frac{V_s}{0.6}\right) = \frac{2}{3}V_s$$

$$m_T = m_s + m_w = \rho_s V_s + \rho_w V_w$$
 but $\rho_w = 1$.

$$m_T = \rho_s V_s + \frac{2}{3} V_s = V_s \left(\rho_s + \frac{2}{3} \right)$$
 and $\rho_{soil} = \frac{m_T}{V_T} = \frac{V_s \left(\rho_s + \frac{2}{3} \right)}{\frac{5}{3} V_s} = \frac{3}{5} \left(\rho_s + \frac{2}{3} \right) = \left(\frac{3\rho_s + 2}{5} \right)$

The mass fraction of SiO₂ in the solids is $f_{SiO_2} = \frac{m_{SiO_2}}{m_s}$ so that $m_{siO_2} = f_{SiO_2} m_s = f_{SiO_2} \rho_s V_s$ thus

$$m_{si} = \left(\frac{M_{Si}}{M_{SiO_2}}\right) m_{SiO_2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}}\right) \rho_s V_s$$

$$F_{Si} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \frac{\rho_s V_s}{V_s \left(\rho_s + \frac{2}{3} \right)} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_s}{\rho_s + \frac{2}{3}} \right)$$

$$F_{Si} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{3\rho_s}{3\rho_s + 2} \right)$$

$$\frac{m_O^{SiO_2}}{m_T} = F_O^{SiO_2} \qquad m_O^{SiO_2} = \left(\frac{2M_O}{M_{SiO_2}}\right) m_{SiO_2} \qquad m_O^{SiO_2} = f_{SiO_2} \left(\frac{2M_O}{M_{SiO_2}}\right) \rho_s V_s$$
Now and

$$F_{O}^{SiO_{2}} = f_{SiO_{2}} \left(\frac{2M_{O}}{M_{SiO_{2}}} \right) \frac{\rho_{s}V_{s}}{V_{s} \left(\rho_{s} + \frac{2}{3} \right)} \quad \text{or} \quad F_{O}^{SiO_{2}} = f_{SiO_{2}} \left(\frac{2M_{O}}{M_{SiO_{2}}} \right) \left(\frac{3\rho_{s}}{3\rho_{s} + 2} \right).$$

$$F_O^{H_2O} = \frac{m_O^{H_2O}}{m_T} = \frac{1}{m_T} \left(\frac{M_O}{M_{H_2O}} \right) m_w$$
 but $m_w = \rho_w V_w$ and $\rho_w = 1$ so that

$$F_O^{H_2O} = \frac{1}{V_s \left(\rho_s + \frac{2}{3}\right)} \left(\frac{M_O}{M_{H_2O}}\right) \frac{2}{3} V_s = \left(\frac{M_O}{M_{H_2O}}\right) \left(\frac{2}{3\rho_s + 2}\right)$$

$$F_H = \left(\frac{2M_H}{M_{H_2O}}\right) \left(\frac{2}{3\rho_s + 2}\right)$$

$$n_H \left(@/B \bullet cm \right) = \frac{F_H \rho_s}{M_H} N_A \times 10^{-24}$$

B-3. CONTAMINATED SATURATED SOIL CALCULATIONS

 V_S = Volume of solids in the porous soil

 V_{SS} = Volume of saturated soil

 V_W = Volume of water in the pores

 V_{CSS} = Volume of contaminated saturated soil

 ρ_S = Mass density of compacted soil

 ρ_{SS} = Mass density of clean saturated soil

 ρ_{CSS} = Mass density of contaminated saturated soil

 ρ_W = Mass density of water (1.0 g/cc)

 ρ_{PuO_2} = Theoretical mass density of PuO₂ (11.46 g/cc)

 m_{SS} = Mass of saturated soil

 m_W = Mass of water in soil

 m_S = Mass of soil solids

 m_{CSS} = Mass of continuated saturated soil

 m_{Si} = Mass of Silicon present

 m_{SiO_2} = Mass of SiO₂ present

 $m_O^{H_2O}$ = Mass of oxygen contributed by H₂O

 M_i = Molecular weight (g/mole) of substance i

 F_{Si} = Mass fraction of Si in CSS

 $F_O^{SiO_2}$ = Mass fraction of oxygen contributed by SiO₂

 f_{SiO_2} = Mass fraction is SiO₂ in the dry soil

 μ = Concentration of PuO₂ in CSS

 ξ = Concentration of Pu in CSS

 N_A = Avogadroes number

 $m_{SS} = \rho_S V_S + \rho_W V_W$ but $\rho_W = 1$, and $V_{SS} = V_S + V_W$, and for 40% porosity we have

$$V_S = 0.6 V_{SS}$$
, $V_W = 0.4 V_{SS}$ thus $V_{SS} = \frac{V_S}{0.6} = \frac{5}{3} V_S$. $V_W = 0.4 V_{SS} = \frac{2}{3} V_S$ so that $m_{SS} = \rho_S V_S + \frac{2}{3} V_S = V_S \left(\rho_S + \frac{2}{3} \right)$

Now
$$\xi \equiv \frac{m_{Pu}}{V_{CSS}}$$
 and $\mu \equiv \frac{m_{PuO_2}}{V_{CSS}}$ and thus $\mu = \left(\frac{M_{PuO_2}}{M_{Pu}}\right)\xi$

$$m_{CSS} = m_{SS} + m_{PuO_2} = m_{SS} + \mu V_{CSS}$$
 and $\rho_{CSS} \equiv \frac{m_{CSS}}{V_{CSS}} = \frac{m_{SS}}{V_{CSS}} + \mu$..

Substitution then gives

$$\rho_{SS} = \left(\frac{3\rho_S + 2}{5}\right) \tag{1}$$

Now
$$V_{CSS} = V_{SS} + V_{PuO_2}$$
, $V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{PuO_2}}$ and $V_{SS} = \frac{5}{3}V_S$. $m_{PuO_2} = \mu V_{CSS}$ thus

 $V_{CSS} = \frac{5}{3}V_s + \frac{\mu V_{CSS}}{\rho_{PuO_2}}$ which can be solved for V_{CSS} and manipulated to give

$$\frac{V_S}{V_{CSS}} = \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right) \tag{2}$$

$$\rho_{CSS} = \frac{m_{SS}}{V_{CSS}} + \mu = \frac{V_S}{V_{CSS}} \left(\rho_S + \frac{2}{3} \right) + \mu$$

or

$$\rho_{\mathit{CSS}} = \mu + \frac{3}{5} \Biggl(1 - \frac{\mu}{\rho_{\mathit{PuO}_2}} \Biggr) \Biggl(\rho_{\mathit{S}} + \frac{2}{3} \Biggr)$$

which reduces to

$$\rho_{CSS} = \mu + \left(1 - \frac{\mu}{\rho_{PuO_2}}\right) \rho_{SS} \tag{3}$$

Now

$$m_{Si} = \left(\frac{M_{Si}}{M_{SiO_2}}\right) m_{SiO_2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}}\right) \rho_S V_S$$

$$F_{Si} = \frac{m_{Si}}{m_{CSS}} , \ \rho_{CSS} V_{CSS} = m_{CSS}$$

so that

$$F_{Si} = \frac{\left(\frac{M_{Si}}{M_{SiO_2}}\right) f_{SiO_2} \rho_S V_S}{\rho_{CSS} V_{CSS}} = \left(\frac{V_S}{V_{CSS}}\right) \frac{\rho_S}{\rho_{CSS}} \left(\frac{M_{Si}}{M_{SiO_2}}\right) f_{SiO_2}$$
thus

$$F_{\mathit{Si}} = \frac{3}{5} \Biggl(1 - \frac{\mu}{\rho_{\mathit{PuO}_2}} \Biggr) \Biggl(\frac{\rho_\mathit{S}}{\rho_\mathit{CSS}} \Biggr) \Biggl(\frac{M_\mathit{Si}}{M_\mathit{SiO_2}} \Biggr) f_{\mathit{SiO}_2}$$

or

$$F_{Si} = \frac{3}{5} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_S}{\rho_{CSS}} \right) f_{SiO_2} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right). \tag{4}$$

Now

$$F_O^{SiO_2} = \frac{3}{5} \left(\frac{2M_O}{M_{SiO_2}} \right) \left(\frac{\rho_S}{\rho_{CSS}} \right) f_{SiO_2} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right). \tag{5}$$

Now consider the water.

$$F_{O}^{H_{2}O} \equiv \frac{m_{O}^{H_{2}O}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{M_{O}}{M_{H_{2}O}} \right) m_{W}$$

but $m_W = \rho_W V_W$ and $\rho_W = 1$ so that $m_W = V_W = \frac{2}{3} V_S$ and $m_{CSS} = \rho_{CSS} V_{CSS}$

$$F_O^{H_2O} = \left(\frac{M_O}{M_{H_2O}}\right) \frac{2}{3} V_S \frac{1}{\rho_{CSS} V_{CSS}} = \left(\frac{V_S}{V_{CSS}}\right) \frac{2}{3\rho_{CSS}} \left(\frac{M_O}{M_{H_2O}}\right)$$

then

$$F_O^{H_2O} = \frac{2}{3\rho_{CSS}} \left(\frac{M_O}{M_{H_2O}} \right) \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$

or finally,

$$F_O^{H_2O} = \frac{2}{5\rho_{CSS}} \left(\frac{M_O}{M_{H_2O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$
 (6)

$$F_{H}^{H_{2}O} = \frac{2}{5\rho_{CSS}} \left(\frac{2M_{H}}{M_{H_{2}O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_{2}}} \right)$$

There are no other hydrogen contributors to the mixture, so we have $F_H = F_H^{H_2O}$.

$$F_{H} = \frac{2}{5\rho_{CSS}} \left(\frac{2M_{H}}{M_{H_{2}O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_{2}}} \right)$$
 (7)

The density of hydrogen in the mixture is $F_{H}\rho_{CSS}$ thus

$$n_H \left(@/B \bullet cm \right) = \frac{F_H \rho_{CSS} N_A}{M_H} \times 10^{-24}$$
(8)

The contribution of the other oxides to the oxygen are calculated and summed to obtain the total oxygen mass fraction. The contribution of the other cations are calculated similarly to the case of silicon. Consider the plutonium oxide.

$$F_{Pu} \equiv \frac{m_{Pu}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}}\right) m_{PuO_2}$$
 but $m_{CSS} = \rho_{CSS} V_{CSS}$ and $m_{PuO_2} = \mu V_{Css}$ so that

substituting gives

$$F_{Pu} = \frac{1}{\rho_{CSS} V_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \mu V_{CSS} = \frac{\mu}{\rho_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right)$$

and thus

$$F_{Pu} = \frac{\mu}{\rho_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \tag{9}$$

$$F_{O}^{PuO_{2}} \equiv \frac{m_{O}^{PuO_{2}}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{2M_{O}}{M_{PuO_{2}}} \right) m_{PuO_{2}} = \frac{1}{\rho_{CSS} V_{CSS}} \left(\frac{2M_{O}}{M_{PuO_{2}}} \right) \mu V_{CSS}$$

so that

$$F_O^{PuO_2} = \left(\frac{\mu}{\rho_{CSS}}\right) \left(\frac{2M_O}{M_{PuO_2}}\right) \tag{10}$$

$$n_{Pu} \left(@/B \bullet cm \right) = \frac{F_{Pu} \rho_{CSS} N_A}{M_{Pu}} \times 10^{-24}$$

Substituting $\mu = 0$ in the above expressions causes them to reduce to the simple forms of the derivation for clean 40% porous saturated soil. For $\mu \to 0$ we have

$$\frac{V_{\scriptscriptstyle S}}{V_{\scriptscriptstyle CSS}} \rightarrow \frac{3}{5} \ , \ \frac{\rho_{\scriptscriptstyle S}}{\rho_{\scriptscriptstyle CSS}} \rightarrow \frac{5\rho_{\scriptscriptstyle S}}{3\rho_{\scriptscriptstyle S}+2} \ , \ F_{\scriptscriptstyle Si} \rightarrow \frac{3}{5} \left(\frac{M_{\scriptscriptstyle Si}}{M_{\scriptscriptstyle SiO_2}}\right) \left(\frac{\rho_{\scriptscriptstyle S}}{\rho_{\scriptscriptstyle CSS}}\right) f_{\scriptscriptstyle SiO_2} \ . \ \text{Substitution gives}$$

$$F_{Si} o rac{3}{5} \left(rac{M_{Si}}{M_{SiO_2}}
ight) f_{SiO_2} rac{5\rho_S}{3\rho_S + 2} = f_{SiO_2} \left(rac{M_{Si}}{M_{SiO_2}}
ight) \left(rac{3\rho_S}{3\rho_S + 2}
ight)$$

which agrees with the previous derivation.

$$F_O^{SiO_2} \to f_{SiO_2} \left(\frac{2M_O}{M_{SiO_2}} \right) \frac{3}{5} \left(\frac{5\rho_S}{3\rho_S + 2} \right) = f_{SiO_2} \left(\frac{2M_O}{M_{SiO_2}} \right) \left(\frac{3\rho_S}{3\rho_S + 2} \right)$$

$$F_{O}^{H_{2}O} \to \frac{2}{5\rho_{CSS}} \left(\frac{M_{O}}{M_{H_{2}O}} \right) = \frac{2}{5} \left(\frac{M_{O}}{M_{H_{2}O}} \right) \frac{5}{3\rho_{S} + 2} = \left(\frac{M_{O}}{M_{H_{2}O}} \right) \left(\frac{2}{3\rho_{S} + 2} \right)$$

$$F_{H} \to \frac{2}{5\rho_{CSS}} \left(\frac{2M_{H}}{M_{H_{2}O}}\right) = \frac{2}{5} \left(\frac{2M_{H}}{M_{H_{2}O}}\right) \frac{5}{3\rho_{S} + 2} = \left(\frac{2M_{H}}{M_{H_{2}O}}\right) \left(\frac{2}{3\rho_{S} + 2}\right)$$

It can be seen that these expressions all agree with the previous derivation.

B-4. WET PUO₂ CALCULATIONS

 ρ_{pwdr} = Packing density of PuO₂ (4.0 g/cc)

 ρ_{PuO_2} = Theoretical PuO₂ density (11.46 g/cc)

 ρ_{mix} = Saturated PuO₂ density

 $m_O^{PuO_2}$ = Mass of oxygen contributed by PuO₂

 $m_O^{H_2O}$ = Mass of oxygen contributed by H₂O

 M_i = Molecular weight of substance i

 m_i = Mass of substance i

 V_i = Volume of substance i

 F_O = Mass fraction of oxygen in the mix

 F_{H} = Mass fraction of hydrogen in the mix

 F_{Pu} = Mass fraction of plutonium in the mix

 F_{PuO_2} = Mass fraction of PuO₂ in the mix

 $F_{H,O}$ = Mass fraction of H₂O in the mix

 n_O = Atomic density of oxygen in the mix (@/ $B \bullet cm$)

 n_H = Atomic density of hydrogen in the mix $(@/B \bullet cm)$

 n_{Pu} = Atomic density of plutonium in the mix $(@/B \bullet cm)$

 n_T = Total atom density in the mix $(n_O + n_H + n_{Pu})$

 N_A = Avogadroes number

$$\rho_{pwdr} = \frac{m_{PuO_2}}{V_{PuO_2}} \qquad V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{PuO_2}} \\ \text{or} \qquad V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{PuO_2}} \\ \text{or} \qquad m_{PuO_2} = \rho_{PuO_2} V_{PuO_2} \\ \text{and} \quad \rho_{pwdr} = \rho_{PuO_2} \left(\frac{V_{PuO_2}}{V_{pwdr}}\right)$$

$$\frac{V_{PuO_2}}{V_{pwdr}} = \frac{\rho_{pwdr}}{\rho_{PuO_2}} = \frac{4.0}{11.46} = 0.3490$$
 and $V_{pwdr} = V_{mix}$

The solid is only 34.9% of the volume. The rest is pore space, to be occupied by water.

$$m_{mix} = \rho_{PuO_2} V_{PuO_2} + \rho_{H_2O} V_{H_2O}$$
 but $\rho_{H_2O} = 1.0$

and

$$V_{PuO_2} = 0.3490 V_{pwdr} = 0.3490 V_{mix}$$
 $V_{H_2O} = 0.6510 V_{pwdr} = 0.6510 V_{mix}$

so that

$$m_{mix} = \rho_{PuO_2} (0.349V_{mix}) + 0.651V_{mix}$$
 $m_{mix} = (0.349\rho_{PuO_2} + 0.651)V_{mix} = 4.6505V_{mix}$

$$\rho_{mix} = \frac{m_{mix}}{V_{mix}} = 4.6505 \text{ g/cc}$$
(1)

Now consider the mass fractions.

$$m_{mix} = m_{PuO_2} + m_{H_2O}$$
 $m_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) m_{PuO_2}$ $m_O^{PuO_2} = \left(\frac{2M_O}{M_{PuO_2}}\right) m_{PuO_2}$

$$m_{H} = \left(\frac{2M_{H}}{M_{H_{2}O}}\right) m_{H_{2}O}$$
 $m_{O}^{H_{2}O} = \left(\frac{M_{O}}{M_{H_{2}O}}\right) m_{H_{2}O}$ $m_{O} = m_{O}^{H_{2}O} + m_{O}^{PuO_{2}}$

$$F_{Pu} = \frac{m_{Pu}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) \frac{m_{PuO_2}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) F_{PuO_2}$$

$$F_{_{\! H}} = \! \left(\frac{2 M_{_{\! H}}}{M_{_{\! H_2O}}} \right) \! \frac{m_{_{\! H_2O}}}{m_{_{\! mix}}} = \! \left(\frac{2 M_{_{\! H}}}{M_{_{\! H_2O}}} \right) \! F_{_{\! H_2O}} \qquad F_{_{\! O}} = \frac{m_{_{\! O}}}{m_{_{\! mix}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! PuO_2}}} \right) \! \frac{m_{_{\! PuO_2}}}{m_{_{\! mix}}} + \! \left(\frac{M_{_{\! O}}}{M_{_{\! H_2O}}} \right) \! \frac{m_{_{\! H_2O}}}{m_{_{\! mix}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! PuO_2}}} \right) \! \frac{m_{_{\! PuO_2}}}{m_{_{\! mix}}} + \! \left(\frac{M_{_{\! O}}}{M_{_{\! H_2O}}} \right) \! \frac{m_{_{\! H_2O}}}{m_{_{\! mix}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! PuO_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! mix}}} + \! \left(\frac{M_{_{\! O}}}{M_{_{\! H_2O}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! mix}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} + \! \left(\frac{M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} + \! \left(\frac{M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}} \right) + \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) \! \frac{m_{_{\! D_2O_2}}}{m_{_{\! D_2O_2}}} = \! \left(\frac{2 M_{_{\! O}}}{M_{_{\! D_2O_2}}} \right) + \! \left(\frac{2 M_{_{\! O}}}{M_$$

$$F_{O} = \left(\frac{2M_{O}}{M_{PuO_{2}}}\right) F_{PuO_{2}} + \left(\frac{M_{O}}{M_{H_{2}O}}\right) F_{H_{2}O}$$

Consider $V_{\rm mix}=1.0$ cc. Then $m_{\rm mix}=\rho_{\rm mix}V_{\rm mix}=4.650~{\rm g}$.

Now we have $V_{PuO_2} = 0.3490 \text{ cc}$, $V_{H_2O} = 0.651 \text{ cc}$ so that

$$m_{PuO_2} = (0.3490)(11.46) = 3.9995 \text{ g}$$
 $m_{H_2O} = (0.6510)(1.0) = 0.6510 \text{ g}$

$$F_{PuO_2} = \frac{m_{PuO_2}}{m_{mix}} = \frac{3.9995}{4.650} = 0.860$$
 $F_{H_2O} = \frac{m_{H_2O}}{m_{mix}} = \frac{0.651}{4.650} = 0.140$

$$\left(\frac{2M_O}{M_{PuO_2}}\right) = \frac{2(15.9995)}{271.0512} = 0.118055 \qquad \left(\frac{M_O}{M_{H_2O}}\right) = \frac{15.9995}{18.01514} = 0.888114$$

$$F_{O} = \left(\frac{2M_{O}}{M_{PuO_{2}}}\right) F_{PuO_{2}} + \left(\frac{M_{O}}{M_{H_{2}O}}\right) F_{H_{2}O} = (0.118055)(0.86) + (0.888114)(0.14) = 0.22586$$

$$F_{Pu} = \left(\frac{239.0522}{271.0512}\right) F_{PuO_2} = 0.75848$$

$$F_H = \left(\frac{2(1.00782)}{18.01514}\right) F_{H_2O} = 0.01566$$

$$n_{Pu}\left(@/B \bullet cm \right) = \frac{m_{Pu}N_A}{V_{mix}M_{Pu}} \times 10^{-24} = \frac{1}{V_{mix}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{M_{Pu}} N_A \times 10^{-24} = \frac{m_{PuO_2}}{V_{mix}} \left(\frac{N_A}{M_{PuO_2}} \right) \times 10^{-24}$$

or
$$n_{Pu} (@/B \bullet cm) = 8.8859 \times 10^{-3}$$

$$n_{H}\left(@/B \bullet cm \right) = \frac{1}{V_{mix}} \left(\frac{2M_{H}}{M_{H,O}} \right) \frac{m_{H_{2}O}}{M_{H}} N_{A} \times 10^{-24} = 4.3523 \times 10^{-2}$$

$$n_O(@/B \bullet cm) = \frac{F_O \rho_{mix}}{M_O} N_A \times 10^{-24} = 3.9537 \times 10^{-2}$$

$$n_T = n_O + n_H + n_{Pu} = 9.19459 \times 10^{-2}$$
 @/ $B \bullet cm$

$$f_o = \frac{n_o}{n_T} = 4.3000 \times 10^{-1}$$

$$f_H = \frac{n_H}{n_T} = 4.7335 \times 10^{-1}$$

$$f_{Pu} = \frac{n_{Pu}}{n_T} = 9.6650 \times 10^{-2}$$
.

B-5. CALCULATIONS FOR POLYETHYLENE IMPREGNATED PUO₂

$$\rho_{pwdr} = 3.9995 \text{ g/cc}$$

$$\rho_{PuO_2} = 11.46 \text{ g/cc}$$

$$\rho_{mix} = impregnated PuO_2 density$$

$$F_{CH_2}$$
 = mass fraction of poly

$$F_{PuO_2}$$
 = mass fraction of PuO_2

$$F_{Pu}$$
 = mass fraction of Pu

$$F_H = \text{mass fraction of } H$$

$$F_O =$$
mass fraction of O

$$F_C =$$
mass fraction of C

$$V_{PuO_2}$$
 = volume of solid PuO_2

$$V_{pwdr} = \text{volume of powdered } PuO_2$$

$$\rho_{pwdr} = \frac{m_{PuO_2}}{V_{PuO_2}}$$
, or $V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{pwdr}}$

$$\frac{V_{PuO_2}}{V_{pwdr}} = \frac{4.0}{11.46} = 0.3490 , \quad V_{mix} = V_{pwdr}$$

$$m_{mix} = \rho_{PuO_2} V_{PuO_2} + \rho_{CH_2} V_{CH_3}$$
 but $\rho_{CH_2} = 0.93$

and
$$V_{PuO_2} = 0.3490 V_{pwdr} = 0.3490 V_{mix}$$
 , .: $V_{CH_2} = 0.6510 V_{mix}$

$$m_{mix} = \rho_{PuO_2} (0.3490V_{mix}) + \rho_{CH_2} (0.6510V_{mix}) = 4.60497V_{mix}$$

Thus
$$\rho_{mix} = \frac{m_{mix}}{V_{mix}} = 4.60497 \text{ g/cc}$$
 (1)

Now consider the mass fractions.

$$m_{mix} = m_{PuO_2} + m_{CH_2}$$
, $m_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) m_{PuO_2}$

$$m_O = \left(\frac{2M_O}{M_{PuO_2}}\right) m_{PuO_2}$$
 and $m_H = \left(\frac{2M_H}{M_{CH_2}}\right) m_{CH_2}$, $m_C = \left(\frac{M_C}{M_{CH_2}}\right) m_{CH_2}$

$$F_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) F_{PuO_2} \ , \ F_H = \left(\frac{2M_H}{M_{CH_2}}\right) F_{CH_2} \ , \ F_O = \left(\frac{2M_O}{M_{PuO_2}}\right) F_{PuO_2}$$

Consider $V_{\rm mix}=1.0~{\rm cc}$, then $m_{\rm mix}=\rho_{\rm mix}V_{\rm mix}=4.60497~{\rm g}$. Now we have

$$V_{PuO_2} = 0.3490V_{mix} = 0.3490 \text{ cc}, \ V_{CH_2} = 0.6510V_{mix} = 0.6510 \text{ cc}$$

$$m_{PuO_2} = (0.3490)(11.46) = 3.9995 \text{ g}, m_{CH_2} = (0.6510)(0.93) = 0.60543 \text{ g}$$

$$F_{PuO_2} = \frac{m_{PuO_2}}{m_{mix}} = \frac{3.9995}{4.60497} = 0.86853$$

$$F_{CH_2} = \frac{m_{CH_2}}{m_{mix}} = \frac{0.60543}{4.60497} = 0.13147$$

$$\left(\frac{2M_{_O}}{M_{_{PuO_2}}}\right) = 0.118055 \; , \quad F_{_O} = \left(\frac{2M_{_O}}{M_{_{PuO_2}}}\right) F_{_{PuO_2}} = 0.10253$$

$$M_C = 12.01115, \ M_H = 1.00782, \ M_{CH_2} = 14.02679$$

$$\left(\frac{2M_{H}}{M_{CH_{2}}}\right) = 0.143699, \quad F_{H} = 0.143699 \\ F_{CH_{2}} = 0.018892$$

$$F_C = \left(\frac{M_C}{M_{CH_2}}\right) F_{CH_2} = 0.11258$$

$$F_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}}\right) F_{PuO_2} = 0.765998$$

$$n_{Pu}\left(@/B.cm \right) = \frac{m_{Pu}N_A}{V_{mix}M_{Pu}} \times 10^{-24} = \frac{m_{PuO_2}}{V_{mix}} \left(\frac{N_A}{M_{PuO_2}} \right) \times 10^{-24} = 8.8859 \times 10^{-3}$$

Now consider the atomic fractions.

$$f_O = \frac{n_O}{n_T} = 1.69843 \times 10^{-1}$$

$$f_H = \frac{n_H}{n_T} = 4.96820 \times 10^{-1}$$

$$f_C = \frac{n_C}{n_T} = 2.48413 \times 10^{-1}$$

$$f_{Pu} = \frac{n_{Pu}}{n_{T}} = 8.49239 \times 10^{-2}$$

B-6. OXIDATION RATE CALCULATIONS PU METAL

Consider a spherical particle of α^{239} Pu with density $\rho = 19.84$ g/cc.

$$A(t) = 4\pi R^2(t)$$
 = surface area of the sphere at time t.

$$R(t) =$$
radius of the sphere at time t .

m = mass of oxidized Pu at time t.

$$A_0 = A(0) = 4\pi R_0^2$$
 where $R_0 = R(0)$

At time t we have

$$\frac{dm}{dt} = \omega A(t) = \omega \left(g / cm^2 \bullet dy \right) A(cm^2) = 4\pi \omega R^2(t)$$

so that

$$dm = 4\pi\omega R^2(t)dt \quad (1)$$

Now
$$m(t) = \frac{4\pi}{3} \rho \left[R_0^3 - R^3(t) \right]$$
 and differentiating this gives

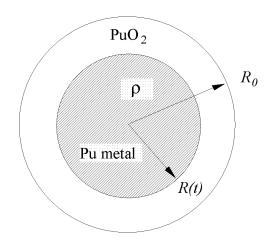
$$dm = \frac{4\pi\rho}{3} \left[-3R^2(t) \frac{dR}{dt} dt \right] = -4\pi\rho R^2(t) \frac{dR(t)}{dt} dt$$

so that we have
$$\frac{dm}{dt} = -4\pi\rho R^2(t) \frac{dR(t)}{dt}$$
 or $dm = -4\pi\rho R^2 dR$.

Substituting in (1) gives $4\pi\omega R^2(t)dt = -4\pi\rho R^2 dR$ or $\omega dt = -\rho dR$.

We then have
$$\int_{R_0}^R dR = -\frac{\omega}{\rho} \int_0^t dt$$
 or $R(t) - R_0 = -\frac{\omega}{\rho} t$. Then $R(t) = R_0 - \frac{\omega}{\rho} t$.

The mass of Pu metal remaining at time t is $M(t) = M_0 - m(t) = \frac{4\pi}{3} \rho R^2(t)$.



Now if we have $6.5 \times 10^{-3} g$ (oxygen) acquired in 900 hours on 1 cm² of surface, we have acquired $\frac{6.5 \times 10^{-3} g}{15.999 g/mol} N_A(@/mol) = 2.44665 \times 10^{20} @ of oxygen. For PuO₂, we have half as many Pu atoms oxidized: <math>n_{Pu} = 1.22332 \times 10^{20} @.$

We then have
$$\frac{1.22332 \times 10^{20} @}{N_A (@/mol)} M_{Pu} (g/mol) = 0.04856 \ g \text{ of Pu} \text{ and}$$

$$\omega = \frac{0.04856 \text{ g/cm}^2}{900 \text{ hrs}} (24 \text{ hrs/dy}) = 1.29495 \times 10^{-3} \frac{\text{g/cm}^2}{\text{dy}}$$

Now consider a 380 gram particle. We have $M_0 = 380 g = \frac{4\pi}{3} \rho R_0^3$.

$$\frac{\omega}{\rho} = \frac{1.29495 \times 10^{-3}}{19.84} = 6.5269 \times 10^{-5} \ cm / dy$$

$$R(t) = 1.65978 \ cm - 6.5269 \times 10^{-5} (cm / dy) t (dy)$$

After 27 years (9855 days), we have R(27yrs) = 1.01655 cm and M(27yrs) = 87.30 g. We thus still have about 23% of the particle mass left. Consider a 2040 g button treated as a sphere. We have

$$M_0 = 2040 \text{ g}, R_0 = 2.90625 \text{ cm}, R(27 \text{yrs}) = 2.26303 \text{ cm}$$

Then M(27yrs) = 963.2 g and we have 47.2% of the mass left.

Now consider the largest particle that would completely oxidize in 27 years.

$$R(27yrs) = 0 = R_0 - 6.5269 \times 10^{-5} cm / dy \cdot t(dy)$$
 or $R_0 = 0.643225 cm$

The mass of the largest spherical particle that would completely oxidize in 27 years is about 22.2 g and is about 0.5 in. in diameter.

The oxidation of plutonium is a complex process that depends on the phase $(\alpha, \beta, \delta, \gamma)$, alloying, oxygen and water availability, and even the prior oxidation exposure of the sample^a. One cannot present a clear picture of the process by merely giving the rate constants over a range of temperatures. In general, high temperatures greatly increase the rate of oxidation, and the presence of water promotes rapid oxidation. Oddly, the presence of oxygen (not in water) retards it. In view of the above, it is impossible to predict how much oxidation of the material has actually occurred. If any of the material is δ phase plutonium, it is likely alloyed with gallium [where does gallium come from?]. In general, δ phase Pu erodes more rapidly than the α phase in water, but alloying with Ga reduces this rate. Hodges, Reynolds, and Haschke^b studied the corrosion of ²³⁹Pu in Rocky Flats tap water and observed that the corrosion rate had two phases: an initial diffusion-limited phase followed by a cracking and flaking phase. The present author made a study²⁰ of the corrosion of Pu spheres using the result of Hodges, et al. The purpose was to determine the rate of contamination of the water surrounding the plutonium in the event of a water flood. A 4.5 kg sphere of α^{239} Pu immersed in water was predicted to produce 43 g of corroded metal in 1350 hours (56.25 days). If the corroded metal were assumed to go to PuO₂, the weight gain of the material would be about 5.76 g of oxygen. The corrosion of δ^{239} Pu was observed to about twice as fast as for α^{239} Pu.

a. ANS, "Plutonium Handbook: A Guide to the Technology", Vol. 1, 1980, Oz Wick, Ed., American Nuclear Society (ANS) La Grange Park, Illinois.

b. Hodges, A. E. III, Reynolds, J., and Haschke, J. M., "Corrosion of Delta Plutonium in Rocky Flats Tap Water", RFP-2891, Rockwell International, Rocky Flats Plant, CO, Sept. 10, 1979 (U) (6pp).

In summary, we can state the following:

More realistic shapes than the assumed sphere will have greater surface area and hence will oxidize more rapidly. If the 380-g-piece considered above were in the shape of a flat sheet, so that area change was almost negligible during the process, it would have oxidized completely by the end of 27 years.

The assumption of a single piece to represent the material is generally conservative. If the material is divided into many pieces, the surface area is greatly increased and again the oxidation will be rapid.

B-7. MCNP CALCULATIONS FOR MINIMUM CRITICAL MASS – PUO₂ AND POLYETHYLENE

A series of computational models were evaluated to determine the minimum critical mass of PuO_2 dispersed in polyethylene. These models evaluated 310 g of ^{239}Pu in the form or PuO_2 combined with varied amounts of polyethylene. The fissile material was dispersed uniformly over the volume of the plutonium-polyethylene sphere. The spherical configuration was fully reflected by a 200-cm-thick sphere of vitrified soil. The hydrogen to plutonium (H/Pu) ratio increased as the amount of polyethylene combined with the fissile material increased. The results of these cases are given in Table B-7.1.

Table B-7.1. 310 g of ^{239}Pu in the form of PuO_2 dispersed in polyethylene in spherical form, fully reflected by 200-cm-thick sphere of vitrified soil, concentration of PuO2 and amount of polyethylene present varied to optimize system.

Radius of Sphere (cm)	Prob. Name	H/Pu Ratio	Sat. Soil Medium Reflector
5	5 cm_310	50.2	0.4423 ± 0.0013
7.5	7-5 cm_310	176.0	0.7345 ± 0.0015
10	10 cm_310	420.9	0.9262 ± 0.0015
12	12 cm_310	729.5	0.9900 ± 0.0013
12.5	12-5 cm_310	824.9	0.9941 ± 0.0013
13	13 cm_310	928.2	0.9957 ± 0.0014
13.5	13-5 cm_310	1039.8	0.9899 ± 0.0013
14	14 cm_310	1159.9	0.9865 ± 0.0012
15	15 cm_310	1427.3	0.9677 ± 0.0012
17.5	17-5 cm_310	2268.2	0.8867 ± 0.0009
20	20 cm_310	3387.1	0.7808 ± 0.0008

Appendix C Composition Calculational Spreadsheets

Appendix C

Composition Calculational Spreadsheets Table C-1. Mixture Calculation Spreadsheet Results

(Since the spreadsheets used will be provided separately, only a few examples will be given.)

Table C-1.1. Pu Contaminated saturated 40% porous soil without ¹⁰B present. (Generated with Spreadsheet CSS2.xls.)

Contaminated Saturated 40% porous Soil

Comp. Dens. (g/cc): PuO2 Dens. (g/cc): Pu conc. (g/cc): Saturated soil dens:	2.38 11.46 0 1.828	MWhydrogen: MWoxygen: MW PuO2: MWH2O:	1.00797 15.9994 271.051 18.01534	Avogadros #: PuO2 conc. (g/cc): Mix dens (g/cc): MWPu:	6.02214E+23 0.000000 1.82800 239.0522
Oxide	MW oxide	MW cation	fi	Fcation (mix)	Foxygen (mix)
SiO2	60.0848	28.086	7.0108E-01	2.5600E-01	2.9167E-01
Al2O3	101.9612	26.9815	1.3272E-01	5.4870E-02	4.8805E-02
Fe2O3	159.6922	55.847	4.7597E-02	2.6006E-02	1.1176E-02
CaO	56.0794	40.08	4.1211E-02	2.3009E-02	9.1847E-03
K2O	94.2034	39.102	3.3482E-02	2.1713E-02	4.4422E-03
MgO	40.3114	24.312	1.9267E-02	9.0773E-03	5.9737E-03
Na2O	61.979	22.9898	1.5348E-02	8.8946E-03	3.0950E-03
TiO2	79.8988	47.9	7.6190E-03	3.5682E-03	2.3837E-03
MnO2	86.9368	54.938	1.1230E-03	5.5437E-04	3.2290E-04
B2O3 (B-11)	70.0238	11.0128	5.6100E-04	1.3785E-04	3.0040E-04
H2O	18.01534	1.00797	not in slag	2.4486E-02	1.9433E-01
PuO2	271.051	239.0522	not in slag	0.0000E+00	0.0000E+00
		Sums:	1.00000	0.42832	0.57168
			Fsum:	1.00000	

Cation	At. No.	ni(@/B.cm)	At. Fraction	Wt. Fraction
Si	14	1.0034E-02	1.2380E-01	2.5600E-01
Al	13	2.2387E-03	2.7621E-02	5.4870E-02
Fe	26	5.1263E-04	6.3249E-03	2.6006E-02
Ca	20	6.3196E-04	7.7972E-03	2.3009E-02
K	19	6.1130E-04	7.5423E-03	2.1713E-02
Mg	12	4.1102E-04	5.0712E-03	9.0773E-03
Na	11	4.2591E-04	5.2549E-03	8.8946E-03
Ti	22	8.2004E-05	1.0118E-03	3.5682E-03
Mn	25	1.1108E-05	1.3706E-04	5.5437E-04
B-11	5	1.3779E-05	1.7001E-04	1.3785E-04
H	1	2.6742E-02	3.2995E-01	2.4486E-02
0	8	3.9335E-02	4.8532E-01	5.7168E-01
Pu-239	94	0	0.0000E+00	0.0000E+00
	Atom Dens.	8.1050E-02	1.0000E+00	1.0000E+00

Calculation treats all boron as B-11

Contaminated Saturated 40% porous Soil

Comp. Dens. (g/cc):	2.38	MWhydrogen:	1.00797	Avogadros #:	6.02214E+23
PuO2 Dens. (g/cc):	11.46	MWoxygen:	15.9994	PuO2 conc. (g/cc):	0.017008
Pu conc. (g/cc):	0.015	MW PuO2:	271.051	Mix dens (g/cc):	1.84229
Saturated soil dens:	1.828	MW H2O:	18.01534	MW Pu:	239.0522
Oxide	MW oxide	MW cation	fi	Feation (mix)	Foxygen (mix)
SiO2	60.0848	28.086	7.0108E-01	2.5364E-01	2.8897E-01
Al2O3	101.9612	26.9815	1.3272E-01	5.4363E-02	4.8354E-02
Fe2O3	159.6922	55.847	4.7597E-02	2.5766E-02	1.1072E-02
CaO	56.0794	40.08	4.1211E-02	2.2796E-02	9.0999E-03
K2O	94.2034	39.102	3.3482E-02	2.1513E-02	4.4012E-03
MgO	40.3114	24.312	1.9267E-02	8.9935E-03	5.9185E-03
Na2O	61.979	22.9898	1.5348E-02	8.8124E-03	3.0664E-03
TiO2	79.8988	47.9	7.6190E-03	3.5352E-03	2.3616E-03
MnO2	86.9368	54.938	1.1230E-03	5.4925E-04	3.1991E-04
B2O3 (B-11)	70.0238	11.0128	5.6100E-04	1.3657E-04	2.9762E-04
H2O	18.01534	1.00797	not in slag	2.4260E-02	1.9254E-01
PuO ₂	271.051	239.0522	not in slag	8.1420E-03	1.0899E-03
		Sums:	1.00000	0.43251	0.56749
			Fsum:	1.00000	

Cation	At. No.	ni(@/B.cm)	At. Fraction	Wt. Fraction
Si	14	1.0019E-02	1.2363E-01	2.5364E-01
Al	13	2.2354E-03	2.7583E-02	5.4363E-02
Fe	26	5.1187E-04	6.3161E-03	2.5766E-02
Ca	20	6.3102E-04	7.7863E-03	2.2796E-02
K	19	6.1039E-04	7.5317E-03	2.1513E-02
Mg	12	4.1041E-04	5.0641E-03	8.9935E-03
Na	11	4.2528E-04	5.2476E-03	8.8124E-03
Ti	22	8.1883E-05	1.0104E-03	3.5352E-03
Mn	25	1.1092E-05	1.3687E-04	5.4925E-04
B-11	5	1.3759E-05	1.6977E-04	1.3657E-04
Н	1	2.6703E-02	3.2949E-01	2.4260E-02
0	8	3.9352E-02	4.8557E-01	5.6749E-01
Pu-239	94	3.77876E-05	4.6627E-04	8.1420E-03
	Atom Dens.	8.1043E-02	1.0000E+00	1.0000E+00

Calculation treats all boron as B-11

Table C-1.2. Pu Contaminated Slag without ¹⁰B present. (Generated with Spreadsheet pusoilm2.xls.) **Dry Compacted and Pu Contaminated Soil**

Comp. Dens. (g/cc):	2.38	MW Pu:	239.0522	Avogadros #:	6.02214E+23
PuO2 dens. (g/cc):	11.46	MWoxygen:	15.9994	PuO2 Conc.(g/cc):	0.000000
Pu conc. (g/cc):	0	MWPuO2:	271.051	Mix dens (g/cc):	2.38000
				Phi:	1.000000

Oxide	MW oxide	MW cation	fi	Fcation (mix)	Foxygen (mix)
SiO2	60.0848	28.086	7.0108E-01	3.2771E-01	3.7337E-01
Al2O3	101.9612	26.9815	1.3272E-01	7.0239E-02	6.2476E-02
Fe2O3	159.6922	55.847	4.7597E-02	3.3291E-02	1.4306E-02
CaO	56.0794	40.08	4.1211E-02	2.9454E-02	1.1757E-02
K2O	94.2034	39.102	3.3482E-02	2.7795E-02	5.6865E-03
MgO	40.3114	24.312	1.9267E-02	1.1620E-02	7.6470E-03
Na2O	61.979	22.9898	1.5348E-02	1.1386E-02	3.9620E-03
TiO2	79.8988	47.9	7.6190E-03	4.5677E-03	3.0513E-03
MnO2	86.9368	54.938	1.1230E-03	7.0966E-04	4.1334E-04
B2O3 (B-11)	70.0238	11.0128	5.6100E-04	1.7646E-04	3.8454E-04
PuO ₂	271.051	239.0522	not in slag	0.0000E+00	0.0000E+00
		Sums:	1.00000	0.51695	0.48305
			Fsum:	1.00000	

Cation	At. No.	ni(@/B.cm)	At. Fraction	Wt. Fraction
Si	14	1.6724E-02	2.4512E-01	3.2771E-01
Al	13	3.7312E-03	5.4687E-02	7.0239E-02
Fe	26	8.5439E-04	1.2523E-02	3.3291E-02
Ca	20	1.0533E-03	1.5438E-02	2.9454E-02
K	19	1.0188E-03	1.4933E-02	2.7795E-02
Mg	12	6.8504E-04	1.0041E-02	1.1620E-02
Na	11	7.0985E-04	1.0404E-02	1.1386E-02
Ti	22	1.3667E-04	2.0032E-03	4.5677E-03
Mn	25	1.8514E-05	2.7136E-04	7.0966E-04
B-11	5	2.2965E-05	3.3660E-04	1.7646E-04
O	8	4.3273E-02	6.3425E-01	4.8305E-01
Pu	94	0.0000E+00	0.0000E+00	0.0000E+00
	Atom Dens	6.8227E-02	1.0000E+00	1.0000E+00

Calculation assumes boron is all B-11.

Dry Compacted and Pu Contaminated Soil

Comp. Dens. (g/cc):	2.38	MW Pu:	239.0522	Avogadros #:	6.02214E+23
PuO2 dens. (g/cc):	11.46	MWoxygen:	15.9994	PuO2 Conc.(g/cc):	0.017008
Pu conc. (g/cc):	0.015	MWPuO2:	271.051	Mix dens (g/cc):	2.39348
_				Phi:	0.992894

Oxide	MW oxide	MW cation	fi	Feation (mix)	Foxygen (mix)
SiO2	60.0848	28.086	7.0108E-01	3.2538E-01	3.7071E-01
Al2O3	101.9612	26.9815	1.3272E-01	6.9740E-02	6.2032E-02
Fe2O3	159.6922	55.847	4.7597E-02	3.3054E-02	1.4204E-02
CaO	56.0794	40.08	4.1211E-02	2.9244E-02	1.1674E-02
K2O	94.2034	39.102	3.3482E-02	2.7598E-02	5.6461E-03
MgO	40.3114	24.312	1.9267E-02	1.1537E-02	7.5926E-03
Na2O	61.979	22.9898	1.5348E-02	1.1305E-02	3.9338E-03
TiO2	79.8988	47.9	7.6190E-03	4.5352E-03	3.0297E-03
MnO2	86.9368	54.938	1.1230E-03	7.0461E-04	4.1041E-04
B2O3 (B-11)	70.0238	11.0128	5.6100E-04	1.7521E-04	3.8181E-04
PuO2	271.051	239.0522	not in slag	6.2670E-03	8.3889E-04
		Sums:	1.00000	0.51954	0.48046
			E	1 00000	

Fsum: 1.00000

Cation	At. No.	ni(@/B.cm)	At. Fraction	Wt. Fraction
Si	14	1.6699E-02	2.4471E-01	3.2538E-01
Al	13	3.7256E-03	5.4596E-02	6.9740E-02
Fe	26	8.5312E-04	1.2502E-02	3.3054E-02
Ca	20	1.0517E-03	1.5412E-02	2.9244E-02
K	19	1.0173E-03	1.4908E-02	2.7598E-02
Mg	12	6.8402E-04	1.0024E-02	1.1537E-02
Na	11	7.0879E-04	1.0387E-02	1.1305E-02
Ti	22	1.3647E-04	1.9999E-03	4.5352E-03
Mn	25	1.8487E-05	2.7091E-04	7.0461E-04
B-11	5	2.2931E-05	3.3604E-04	1.7521E-04
0	8	4.3284E-02	6.3430E-01	4.8046E-01
Pu	94	3.7788E-05	5.5375E-04	6.2670E-03
	Atom Dens.	6.8239E-02	1.0000E+00	1.0000E+00

Calculation assumes boron is all B-11.

Appendix D Typical MCNP Input Listings

Appendix D

Typical MCNP Input Listings

D-1. MCNP LISTING FOR INFINITE X INFINITE X 14 FT THICK VITRIFIED SOIL (70.1 WT% SIO₂ IN SOIL) AND PUO₂, FULL REFLECTION ON TOP AND BOTTOM.

```
U05-02.i 14 ft (426.7 cm) thick infinite slab of PuO2 and
    compacted soil sandwiched between 4 ft (121.92 cm)
    of compacted soil on the bottom and 6 ft (182.88 cm)
    of uncompacted soil on the top. All soil is dry.
    Concentration = 0.0025 g Pu/cc of mix. Periodic
    boundary conditions are used.
                      -5 6 -7 8 -3 4 imp:n=1 $ Vitrified bottom soil 
-5 6 -7 8 -2 3 imp:n=1 $ PuO2 & vitr soil
        -2.38
        -2.38225
                      -5 6 -7 8 -2 3 IIIIp.II-1
-5 6 -7 8 -1 2 imp:n=1
                                                  $ Top soil
3
    3
        -1.43
                      1:-4:5:-6:7:-8
                                         imp:n=0 $ elsewhere
C
    Surface Cards
      pz 609.60
      pz 426.72
3
      pz 0.
      pz -121.92
5 -6 px 100.
6 -5 px -100.
7 -8 py 100.
8 -7 py -100.
    Data Cards
    Clean vitrified soil
     14000.50c 2.4512e-1 13027.50c 5.4687e-2
     26000.55c 1.2523e-2
                            20000.50c
                                        1.5438e-2
     19000.50c 1.4933e-2 12000.50c
                                        1.0041<del>e</del>-2
     11023.50c 1.0404e-2
                           22000.50c 2.0032<del>e</del>3
     25055.50c 2.7136e-4
      5011.50c 3.3660e-4 8016.50c 6.3425e-1
    vitrified soil mixed with PuO2 (0.0025 g Pu/cc)
C
     14000.50c 2.4505e-1 13027.50c 5.4672e-2
                                        1.5433e-2
     26000.55c 1.2519e-2 20000.50c
                            12000.50c
     19000.50c 1.4929e-2
11023.50c 1.0401e-2
                                        1.0038e-2
                                        2.0027e-3
                            22000.50c
     25055.50c
                 2.7129e-4
      5011.50c 3.3651e-4
                              8016.50c 6.3426e-1
     94239.50c 9.2306e-5
C
    Clean dry soil
     14000.50c 2.4512e-1 13027.50c
                                        5.4687e-2
     26000.55c 1.2523e-2 20000.50c
                                        1.5438<del>e</del> 2
     19000.50c 1.4933e-2 12000.50c
                                        1.0041e-2
     11023.50c 1.0404e-2
                            22000.50c 2.0032e-3
     25055.50c 2.7136e-4
      5011.50c 3.3660e-4
                            8016.50c 6.3425e-1
kcode 1000 1.0 15 115
```

D-2. MCNP LISTING FOR INFINITE X INFINITE X 14 FT THICK VITRIFIED SOIL (80 WT% SIO₂ IN SOIL) AND PUO₂, FULL REFLECTION ON TOP AND BOTTOM.

```
U04-06.i 14 ft (426.7 cm) thick infinite slab of PuO2 and
    compacted soil sandwiched between 4 ft (121.92 cm)
    of compacted soil on the bottom and 6 ft (182.88 cm)
    of uncompacted soil on the top. All soil is dry.
    Concentration = 0.002 g Pu/cc of mix. Periodic
    1:-4:5:-6:7:-8
                                            imp:n=0 $ elsewhere
    Surface Cards
      pz 609.60
2
       pz 426.72
3
       pz 0.
4
      pz -121.92
5
      px 100.
 -6
6 -5
      px -100.
      py 100.
py -100.
7 -8
8 -7
    Data Cards
C
C
    Clean vitrified soil (80 wt% SiO2)
      14000.50c 2.7525e-1 13027.50c 3.6010e-2
      26000.55c 8.2454e-3 20000.50c
                                          1.0165e-2
      19000.50c 9.8325e3 12000.50c 6.6113e3
      11023.50c 6.8503e3 22000.50c 1.3190e3
      25055.50c 1.7867e-4
       5011.50c 2.2164e4 8016.50c 6.4532e-1
C
    vitrified soil mixed with PuO2 (0.002 g Pu/cc)(80 wt% SiO2)

    14000.50c
    2.7519e1
    13027.50c
    3.6002e2

    26000.55c
    8.2436e3
    20000.50c
    1.0162e2

    19000.50c
    9.8304e3
    12000.50c
    6.6099e3

    11023.50c
    6.8488e3
    22000.50c
    1.3187e3

                                          1.3187e-3
      25055.50c 1.7864e-4
       5011.50c 2.2159e-4
                               8016.50c 6.4533e-1
      94239.50c 7.2669e-5
    Clean dry soil
                        (80 wt% SiO2)
      14000.50c 2.7525e1 13027.50c 3.6010e2
      26000.55c 8.2454e-3
                             20000.50c
                                          1.0165<del>e</del>-2
      19000.50c 9.8325e-3
                             12000.50c 6.6113e-3
      11023.50c 6.8503e-3 22000.50c 1.3190e-3
25055.50c 1.7867e4
5011.50c 2.2164e4 8016.50c 6.4532e1
kcode 1000 1.0 15 115
ksrc 0 0 150.
```

D-3. MCNP LISTING FOR INFINITE X INFINITE X 14 FT THICK VITRIFIED SOIL (90 WT% SIO₂ IN SOIL) AND PUO₂, FULL REFLECTION ON TOP AND BOTTOM

```
U03-08.i 14 ft (426.7 cm) thick infinite slab of PuO2 and
    compacted soil sandwiched between 4 ft (121.92 cm)
    of compacted soil on the bottom and 6 ft (182.88 cm)
    of uncompacted soil on the top. All sil is dry.
    Concentration = 0.0015 g Pu/cc of mix. Periodic
    boundary conditions are used. (Soil 90 wt% SiO2)
        -2.38
                      -5 6 -7 8 -3 4 imp:n=1 $ Vitrified bottom soil
                      -5 6 -7 8 -2 3 imp:n=1 $ PuO2 & vitr soil
-5 6 -7 8 -1 2 imp:n=1 $ Top soil
        -2.38135
3
    3
        -1.43
4
                       1:-4:5:-6:7:-8
                                         imp:n=0 $ elsewhere
C
    Surface Cards
      pz 609.60
      pz 426.72
3
      pz 0.
      pz -121.92
5 -6 px 100.
6 -5 px -100.
7 -8 py 100.
8 -7 py -100.
    Data Cards
C
    Clean vitrified soil (90 wt% SiO2)
     14000.50c 3.0475e-1 13027.50c 1.7717e-2
     26000.55c  4.0568e3  20000.50c  5.0012e3
19000.50c  4.8378e3  12000.50c  3.2528e3
11023.50c  3.3706e3  22000.50c  6.4898e4
     11023.50c 3.3706e-3
25055.50c 8.7910e-5
      5011.50c 1.0904e-4 8016.50c
                                          6.5616e-1
    vitrified soil mixed with PuO2 (0.0015 g Pu/cc)(Soil 90 wt% SiO2)
     14000.50c 3.0470e-1 13027.50c 1.7715e-2
     26000.55c 4.0562e-3 20000.50c
                                         5.0004e-3
     19000.50c 4.8371e-3 12000.50c 3.2522e-3
     11023.50c 3.3700e-3 22000.50c 6.4888e-4
     25055.50c 8.7896<del>e</del>5
      5011.50c 1.0903e-4
                             8016.50c 6.5617<del>e</del>1
     94239.50c 5.3640e-5
    Clean dry soil
                       (90 wt% SiO2)
C
     14000.50c 3.0475e1 13027.50c
26000.55c 4.0568e3 20000.50c
                                         1.7717e-2
                                          5.0012e-3
                            19000.50c 4.8378<del>e</del>3
                                          3.2528e-3
     11023.50c 3.3706e-3
                             22000.50c
                                         6.4898e-4
     25055.50c 8.7910e-5
      5011.50c 1.0904e-4 8016.50c
                                          6.5616e-1
kcode 1000 1.0 15 115
ksrc
      0 0 150.
```

D-4. MCNP LISTING FOR INFINITE X INFINITE X 14 FT THICK VITRIFIED SOIL (100 WT% SIO₂ IN SOIL) AND PUO₂, FULL REFLECTION ON TOP AND BOTTOM

```
U02-10.i 14 ft (426.7 cm) thick infinite slab of PuO2 and
     compacted soil sandwiched between 4 ft (121.9 cm)
     of compacted soil on the bottom and 6 ft (182.88 cm)
     of uncompacted soil on the top. All soil is dry.
     Concentration = 0.001 \text{ g Pu/cc} of mix. Periodic
     boundary conditions are used. ("Soil" in slab is 100 wt% SiO2)
         -2.38
                         -5 6 -7 8 -3 4 imp:n=1 $ Vitrified bottom soil
                         -5 6 -7 8 -2 3 imp:n=1 $ PuO2 & vitr soil
-5 6 -7 8 -1 2 imp:n=1 $ Top soil
          -2.38090
3
     3
         -1.43
4
                          1:-4:5:-6:7:-8
                                               imp:n=0 $ elsewhere
C
     Surface Cards
       pz 609.60
       pz 426.72
3
       pz 0.
       pz -121.92
5 -6 px 100.
6 -5 px -100.
7 -8 py 100.
8 -7 py -100.
     Data Cards
C
     Clean vitrified soil
      14000.50c 2.4512e1 13027.50c 5.4687e2 26000.55c 1.2523e2 20000.50c 1.543&e-2 19000.50c 1.4933e2 12000.50c 1.0041e2 11023.50c 1.0404e2 22000.50c 2.0032e3 25055.50c 2.7136e4 5011.50c 3.3660e4 8016.50c 6.3425e1
C
     100 wt% SiO2 mixed with PuO2 (0.001 g Pu/cc)
      14000.50c 3.3330e-1
       8016.50c 6.6667e-1
      94239.50c 3.5202e-5
C
     Clean dry soil
      14000.50c 2.4512e-1 13027.50c 5.4687e-2
      26000.55c 1.2523e-2 20000.50c 1.5438e-2
      19000.50c 1.4933e-2 12000.50c 1.0041e-2
11023.50c 1.0404e-2 22000.50c 2.0032e-3
25055.50c 2.7136e-4
5011.50c 3.3660e-4 8016.50c 6.3425e-1
kcode 1000 1.0 15 115
ksrc 0 0 150.
```

D-5. MCNP LISTING FOR 30 FT X 30 FT X 14 FT THICK VITRIFIED SOIL (70.1 WT% SIO₂ IN SOIL) AND PUO₂, FULL REFLECTION ON TOP, HORIZONTAL SIDES, AND BOTTOM

```
U05-02 fin4 \, 14 ft (426.7 cm) thick x 30 ft x 30 ft slab of PuO2 and
    vitrified soil sandwiched between 4 ft (121.92 cm)
    of vitrified soil on the bottom and 6 ft (182.88 cm)
    of uncompacted soil on the top. All soil is dry.
    Concentration = 0.0025 g Pu/cc of mix.
    ("Soil" in slab is 70.1 wt% SiO2)
        -2.38225
                   -5 +6 -7 +8 +3 -2
                                           u=1 imp:n=1
                                                                $ PuO2 & vitr soil
        -2.38
                     (+5:-6:+7:-8) +3 -2 u=1 imp:n=1
                                                                $ Vitrified soil
3
    1
        -2.38
                     - 3
                                           u=1 imp:n=1
                                                                $ Vitrified soil
    3
        -1.43
                     +2
                                            u=1 imp:n=1
                                                                 $ Top soil
5
    0
                     -9 +10 -11 +12 -1 +4 fill=1 imp:n=1 $ Boundary
6
                     +9:-10:+11:-12:+1:-4
                                                    imp:n=0 $ ziow
    Surface Cards
      pz 609.60
2
      pz 426.72
3
      pz 0.
      pz -121.92
4
      px 457.20
5
      px -457.20
6
7
      py 457.20
8
      py -457.20
9
      px 609.60
10
      px -609.60
      py 609.60
11
      py -609.60
12
      pz -304.80
pz 792.48
13
14
C
    Data Cards
    Clean vitrified soil
C.
     14000.50c 2.4512e-1 13027.50c 5.4687e-2
     26000.55c 1.2523e-2 20000.50c
                                        1.5438e-2
     19000.50c 1.4933e-2
                           12000.50c
                                        1.0041<del>e</del>2
     11023.50c 1.0404e-2 22000.50c 2.0032e-3
     25055.50c 2.7136e-4
      5011.50c 3.3660e-4 8016.50c 6.3425e-1
    vitrified soil mixed with PuO2 (0.0025 g Pu/cc)
C
     14000.50c 2.4505e1 13027.50c 5.4672e2
26000.55c 1.2519e2 20000.50c 1.5433e2
19000.50c 1.4929e2 12000.50c 1.0038e2
                            12000.50c
     11023.50c 1.0401e-2
                            22000.50c 2.0027e-3
     25055.50c 2.7129e-4
      5011.50c
                 3.3651e-4
                              8016.50c 6.3426<del>e</del>1
     94239.50c 9.2306e-5
C
    Clean dry soil
     14000.50c 2.4512e-1 13027.50c 5.4687e-2
     26000.55c 1.2523e-2 20000.50c
                                        1.5438e-2
     19000.50c 1.4933e-2
                            12000.50c 1.0041<del>e</del>2
     11023.50c 1.0404e-2
                             22000.50c 2.0032e-3
      5055.50c 2.7136e-4
5011.50c 3.3660e-4
     25055.50c
                             8016.50c 6.3425e-1
kcode 2000 1.0 15 300
ksrc 0 0 150.
```

D-6. MCNP LISTING FOR SPHERE OF VITRIFIED SOIL (70.1 WT% SIO₂ IN SOIL) AND 1200 G OF PUO₂, FULL REFLECTION AROUND SPHERE

```
Y05-01 Sphere of PuO2 and slag with 1200 g Pu.
    Concentration = 0.05 \text{ g Pu/cc} of mix. Sphere
     is surrounded by 5 ft thick sphere of uncompacted
    water saturated soil.
    1 6.8267e-2 -1
                               imp:n=1
    2 8.1050e-2
                    1 - 2 imp:n=1
3
                       2
                               imp:n=0
C
    Surface Cards
    so 17.8940
so 170.294
1
    Data Cards
    vitrified soil mixed with PuO2 (0.05 g Pu/cc)
     14000.50c 2.4376e1 13027.50c 5.4384e-2 26000.55c 1.2453e2 20000.50c 1.5352e-2
      19000.50c 1.4850e-2 12000.50c 9.9850e-3
      11023.50c 1.0347e-2 22000.50c 1.9921e-3
      25055.50c 2.6986e-4
       5011.50c 3.3474e-4
                                8016.50c 6.3443e-1
      94239.50c 1.8451e-3
C
    Clean saturated soil
      14000.50c 1.2380e-1 13027.50c 2.7621e-2
     26000.55c 6.3249e-3 20000.50c
19000.50c 7.5423e-3 12000.50c
11023.50c 5.2549e-3 22000.50c
25055.50c 1.3706e-4
5011.50c 1.7001e-4 1001.50c
                                            7.7972e-3
                               12000.50c
                                            5.0712e-3
                               22000.50c 1.0118e-3
                                 1001.50c 3.2995<del>e</del>1
       8016.50c 4.8532e-1
mt2 lwtr
kcode 1000 1.0 15 115
ksrc 0 0 0
```

D-7. MCNP LISTING FOR SPHERE OF SATURATED SOIL (70.1 WT% SIO₂ AND 40% VOLUME FRACTION IN SOIL CONTAINING H₂O) AND 1200 G OF PUO₂, FULL REFLECTION AROUND SPHERE

```
Z02-01
Sphere of PuO2 and Saturated soil with 1200 g Pu.
    Concentration = 0.020 g Pu/cc of mix. Sphere
    is surrounded by 5 ft thick sphere of uncompacted
C
    water saturated soil.
    1 8.1040e-2 -1
                             imp:n=1
    2 8.1050e-2 1
                        -2 imp:n=1
                              imp:n=0
C
   Surface Cards
   so 24.2859
so 176.686
1
2
    Data Cards
    PuO2 & saturated soil (0.020 g Pu/cc)
14000.50c 1.2357e-1 13027.50c 2.7570e-2
26000.55c 6.3131e-3 20000.50c 7.7826e-3
     19000.50c 7.5282e-3 12000.50c
                                          5.0618<del>c</del>3
     11023.50c 5.2451e-3 22000.50c 1.0099e-3
     25055.50c 1.3680e-4
       5011.50c 1.6969e-4
                              1001.50c 3.2933e-1
       8016.50c 4.8566e-1 94239.50c 6.2171e-4
mt1 lwtr
C
    Clean saturated soil
     14000.50c 1.2380e-1 13027.50c 2.7621e-2
                                          7.7972e-3
     26000.55c 6.3249e-3 20000.50c
     19000.50c 7.5423e3
11023.50c 5.2549e3
25055.50c 1.3706e4
                                          5.0712e-3
                              12000.50c
                              22000.50c 1.0118e-3
       5011.50c 1.7001e-4
                               1001.50c 3.2995<del>e</del>1
       8016.50c 4.8532e-1
mt2 lwtr
kcode 1000 1.0 15 115
ksrc 0 0 0
```

D-8. MCNP LISTING FOR SPHERES OF SATURATED SOIL (70.1 WT% SIO₂ AND 40% VOLUME FRACTION IN SOIL CONTAINING H₂O) AND VARIED AMOUNTS OF PUO₂, SPHERE ARRANGED IN CLOSE PACK CONFIGURATION TO SIMULATED ARRAY OF DRUMS

```
W02-01 Array of spheres of PuO2 and saturated soil.
    Mix Concentration = 0.020 g Pu/cc. The spheres
    are surrounded by saturated soil. The array is 11 \times 11 \times 3 cells high. Four cetral
    cells are abnormal. One has 1200 g Pu and the
    other three have 300 g Pu. the four spheres in
    these cells are clustered together. Normal cells
    have 200 g Pu.
    Contents of lattice cell [0,0,0] (universe 1)
         8.1040e-2
                                        u=1 imp:n=1 $ PuSat. Soil
                   - 5
         8.1050e-2
                     5
                                        u=1 imp:n=1 $ Sat. soil
    Description of lattice cell [0,0,0] and lattice (universe 2)
        -1 2 -3 4 -6 7 lat=1 u=2 imp:n=1 fill=5:5 -5:5 -1:1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1
             1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 5 6 1 1 1 1
         1 1 1 1 1 3 4 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
         1 1 1 1 1 1 1 1 1 1 1
    Description of window containing 11 X 11 X 3 drums
       -8 9 -10 11 -12 13 fill=2 imp:n=1
    Description of outside of drum array
         8:-9:10:-11:12:-13
                               imp:n=0
    Cell with 1200 g Pu (Universe 3)
         8.1040e-2
                   -14
                         u=3 imp:n=1
         8.1050e-2 14
                         u=3 imp:n=1
   Cell with 300 g Pu (Universe 4)
         8.1040e-2 -15 u=4 imp:n=1
```

```
8.1050e-2 15
                          u=4 imp:n=1
    Cell with 300 g Pu
                         (Universe 5)
10
   1
         8.1040e-2 -16
                          u=5 imp:n=1
11
         8.1050e-2
                   16
                          u=5 imp:n=1
C
    Cell with 300 g Pu
                         (Universe 6)
         8.1040e-2 -17
                          u=6 imp:n=1
12
    1
13
         8.1050e-2
                    17
                          u=6 imp:n=1
    Surface Cards
C
    Unit Cell Surfaces
    px 28.63
1
    px -28.63
2
    py 28.63
    py -28.63
4
5
    so 13.3650
6
    pz 41.69
7
    pz -41.69
C
    Window Boundaries
8
    px 314.92
    px -314.92
9
10
    py 314.92
    py -314.92
pz 125.04
11
12
    pz -125.04
13
    Off Normal Sphere Surfaces
C
        4.3441
                -4.3441 0. 24.2859 $ 1200 g sphere
14
    S
15
    s -13.3308
               -13.3308 0. 15.2992 $ 300 g sphere
16
    s 13.3308
               13.3308 0. 15.2992 $ 300 g sphere
                                     $ 300 g sphere
17
    s -13.3308
                 13.3308 0. 15.2992
C
    Data Cards
C
    PuO2 & saturated soil (0.020 g Pu/cc)
     14000.50c 1.2357e-1
                           13027.50c 2.7570e-2
     26000.55c
                6.3131e-3
                           20000.50c
                                     7.7826e-3
                7.5282e-3
                           12000.50c
     19000.50c
                                     5.0618e-3
     11023.50c
                           22000.50c
                                     1.0099e-3
               5.2451<del>e</del>3
     25055.50c
               1.3680e-4
                           1001.50c
      5011.50c
               1.6969e-4
                                     3.2933e-1
      8016.50c 4.8566e-1 94239.50c
                                     6.2171e-4
mt1
    lwtr
C
    Clean saturated soil
                                     2.7621e-2
                          13027.50c
m2
     14000.50c 1.2380e-1
                          20000.50c
                                     7.7972e-3
     26000.55c
               6.3249e-3
     19000.50c
               7.5423e-3
                           12000.50c
                                     5.0712e-3
                5.2549e-3
                           22000.50c
     11023.50c
                                     1.0118e-3
     25055.50c
                1.3706e-4
               1.7001e-4
                            1001.50c 3.2995e-1
      5011.50c
      8016.50c 4.8532e-1
mt2 lwtr
kcode 3000 1.0 15 115
ksrc
        4.3441
                 52.9159 0. $ sphere 14
       43.9292
                 43.9292 0. $ sphere 15
       13.3308
                 13.3308 0. $ sphere 16
       43.9292
                 13.3308 0. $ sphere 17
     -286.30 286.30 -83.38
                                         286.30 -83.38 -171.78 286.30 -83.38
                              -229.04
     -114.52
             286.30 -83.38
                               -57.26
                                         286.30 -83.38
                                                         0. 286.30 -83.38
       57.26
              286.30 -83.38
                                114.52
                                         286.30 -83.38
                                                         171.78 286.30 -83.38
      229.04
              286.30
                     -83.38
                                286.30
                                         286.30 -83.38
                                         229.04 -83.38
                                                        -171.78 229.04 -83.38
     -286.30
              229.04
                     -83.38
                               -229.04
                               -57.26
                                                          0. 229.04 -83.38
     -114.52
              229.04
                     -83.38
                                         229.04 -83.38
       57.26
              229.04 -83.38
                                114.52
                                         229.04 -83.38
                                                         171.78 229.04 -83.38
      229.04
             229.04 -83.38
                                286.30
                                         229.04 -83.38
                               -229.04
                                         171.78 -83.38
                                                        -171.78 171.78 -83.38
     -286.30
             171.78 -83.38
     -114.52 171.78 -83.38
                               -57.26
                                         171.78 -83.38
                                                          0. 171.78 -83.38
       57.26 171.78 -83.38
                                114.52
                                         171.78 -83.38
                                                        171.78 171.78 -83.38
```

```
171.78 -83.38
                           286.30
229.04
                                    171.78 -83.38
-286.30
         114.52 -83.38
                          -229.04
                                    114.52 -83.38
                                                   -171.78 114.52 -83.38
-114.52
         114.52 -83.38
                          -57.26
                                    114.52 -83.38
                                                     0. 114.52 -83.38
  57.26
         114.52 -83.38
                           114.52
                                    114.52 -83.38
                                                    171.78 114.52 -83.38
         114.52 -83.38
                           286.30
                                    114.52 -83.38
229.04
-286.30
          57.26 -83.38
                          -229.04
                                     57.26 -83.38
                                                             57.26 -83.38
                                                   -171.78
                -83.38
                           -57.26
                                     57.26 -83.38
-114.52
          57.26
                                                     0. 57.26 -83.38
          57.26 -83.38
57.26 -83.38
                                     57.26 -83.38
  57.26
                           114.52
                                                     171.78
                                                             57.26 -83.38
 229.04
                           286.30
                                     57.26 -83.38
-286.30
          0. -83.38
                     -229.04
                                  0. -83.38 -171.78
                                                         0. -83.38
          0. -83.38
                      -57.26
                                  0. -83.38
-114.52
                                              0. 0. -83.38
 57.26
          0. -83.38
                       114.52
                                  0. -83.38
                                              171.78
                                                         0. -83.38
229.04
          0. -83.38
                       286.30
                                  0. -83.38
-286.30
         -57.26 -83.38
                          -229.04
                                   -57.26 -83.38
                                                   -171.78 -57.26 -83.38
-114.52
         -57.26
                -83.38
                          -57.26
                                    -57.26 -83.38
                                                     0. -57.26 -83.38
  57.26
         -57.26 -83.38
                           114.52
                                    -57.26 -83.38
                                                    171.78 -57.26 -83.38
229.04
         -57.26 -83.38
                           286.30
                                   -57.26 -83.38
-286.30
        -114.52 -83.38
                          -229.04
                                   -114.52 -83.38
                                                   -171.78 -114.52 -83.38
         -114.52 -83.38
                           -57.26
                                   -114.52 -83.38
-114.52
                                                    0. -114.52 -83.38
  57.26
        -114.52 -83.38
                           114.52
                                   -114.52 -83.38
                                                     171.78 - 114.52 - 83.38
                                   -114.52 -83.38
 229.04
         -114.52 -83.38
                           286.30
                                   -171.78 -83.38
        -114.52 -83.38
-286.30
                          -229.04
                                                    -171.78 -171.78 -83.38
-114.52
         -171.78 -83.38
                          -57.26
                                   -171.78 -83.38
                                                     0. -114.52 -83.38
  57.26
        -171.78 -83.38
                           114.52
                                   -171.78 -83.38
                                                     171.78 -171.78 -83.38
229.04
        -171.78 -83.38
                           286.30
                                   -171.78 -83.38
        -114.52 -83.38
                          -229.04
                                   -229.04 -83.38
-286.30
                                                    -171.78 -229.04 -83.38
-114.52
         -229.04 -83.38
                          -57.26
                                   -229.04 -83.38
                                                     0. -114.52 -83.38
        -229.04 -83.38
                           114.52
                                                     171.78 - 229.04 - 83.38
  57.26
                                   -229.04 -83.38
        -229.04 -83.38
                           286.30
                                  -229.04 -83.38
229.04
-286.30
        -114.52 -83.38
                         -229.04
                                   -286.30 -83.38
                                                   -171.78 -286.30 -83.38
         -286.30 -83.38
                           -57.26
                                   -286.30 -83.38
-114.52
                                                     0. -114.52 -83.38
                                  -286.30 -83.38
                                                     171.78 -286.30 -83.38
 57.26
         -286.30 -83.38
                           114.52
 229.04
         -286.30 -83.38
                           286.30 -286.30 -83.38
          286.30 0. -229.04
                               286.30
-286.30
                                        0. -171.78
                                                      286.30
                                                               0.
                  0. -57.26
                                       0. 0. 286.30 0.
-114.52
          286.30
                              286.30
 57.26
          286.30
                  0. 114.52
                              286.30
                                        0. 171.78
                                                    286.30
                                                              0.
229.04
          286.30
                  0. 286.30
                               286.30
                                        0.
                  0. -229.04
-286.30
          229.04
                               229.04
                                        0. -171.78
                                                      229.04
                                                               0.
                                                       0.
-114.52
          229.04
                  0. -57.26
                              229.04
                                        0. 0. 229.04
  57.26
          229.04
                  0. 114.52
                              229.04
                                        0. 171.78
                                                    229.04
                                                              0.
                  0. 286.30
          229.04
                               229.04
                                        0.
229.04
                  0. -229.04
-286.30
          171.78
                               171.78
                                        0. -171.78
                                                     171.78
                                                               0.
-114.52
          171.78
                  0. -57.26
                              171.78
                                        0. 0. 171.78
                                                       0.
                  0. 114.52
0. 286.30
                                        0. 171.78
          171.78
                              171.78
  57.26
                                                    171.78
                                                              0.
229.04
          171.78
                               171.78
                                        0.
-286.30
          114.52
                  0. -229.04
                               114.52
                                        0. -171.78
                                                     114.52
                                                               0.
                  0. -57.26
-114.52
          114.52
                              114.52
                                        0. 0. 114.52
                                                     0.
                                        0. 171.78
 57.26
          114.52
                  0. 114.52
                              114.52
                                                              0.
                                                  114.52
229.04
          114.52
                  0. 286.30
                              114.52
                                        0.
                  0. -229.04
-286.30
          57.26
                               57.26
                                        0. -171.78
                                                       57.26
                                                               0.
                                                      0.
-114.52
          57.26
                  0. -57.26
                               57.26
                                        0. 0. 57.26
  57.26
          57.26
                  0.114.52
                               57.26
                                        0. 171.78
                                                     57.26
229.04
          57.26
                  0. 286.30
                               57.26
                                        0.
          0. 0. -229.04
                           0. 0. -171.78
-286.30
                                              0.0.
-114.52
          0. 0. -57.26
                           0. 0. 0. 0. 0.
  57.26
          0. 0. 114.52
                           0. 0. 171.78
                                             0.0.
229.04
          0. 0. 286.30
                           0.0.
-286.30
         -57.26
                  0. -229.04
                              -57.26
                                        0. -171.78
                                                     -57.26
                                                                0.
         -57.26
                  0. -57.26
                              -57.26
-114.52
                                       0. 0. -57.26
                                                     0.
                  0. 114.52
 57.26
        -57.26
                              -57.26
                                       0. 171.78
                                                  -57.26
                                                              0.
229.04
        -57.26
                  0. 286.30
                              -57.26
                                       0.
                  0. -229.04 -114.52
                                        0. -171.78 -114.52
-286.30
        -114.52
                                                                0.
-114.52
        -114.52
                  0. -57.26 -114.52
                                       0. 0. -114.52
                                                      0.
  57.26
        -114.52
                  0. 114.52 -114.52
                                       0. 171.78 -114.52
```

```
-114.52 0. 286.30 -114.52
229.04
-286.30
        -114.52
                 0. -229.04 -171.78
                                      0. -171.78 -171.78
-114.52
        -171.78
                 0. -57.26 -171.78
                                      0. 0. -114.52 0.
        -171.78
  57.26
                 0. 114.52 -171.78
                                      0. 171.78 -171.78
        -171.78
                 0. 286.30 -171.78
229.04
                                      0.
-286.30
        -114.52
                 0. -229.04 -229.04
                                      0. -171.78 -229.04
                                                             0.
        -229.04
                           -229.04
-114.52
                 0. -57.26
                                      0. 0. -114.52 0.
 57.26
                 0. 114.52
0. 286.30
        -229.04
                           -229.04
                                      0. 171.78 -229.04
        -229.04
 229.04
                           -229.04
                                      0.
        -114.52
-286.30
                 0. -229.04 -286.30
                                      0. -171.78 -286.30
                                                             Θ.
        -286.30
                 0. -57.26 -286.30
                                      0. 0. -114.52 0.
-114.52
                 0. 114.52
 57.26
        -286.30
                            286.30
                                     0. 171.78 -286.30
                                                           0.
229.04
        -286.30
                 0. 286.30 -286.30
                                      0.
-286.30
         286.30
                 83.38
                        -229.04
                                   286.30 83.38 -171.78
                                                           286.30
                                                                    83.38
-114.52
         286.30
                 83.38
                         -57.26
                                   286.30 83.38
                                                   0. 286.30 83.38
         286.30
  57.26
                 83.38
                          114.52
                                   286.30 83.38
                                                  171.78
                                                           286.30
                                                                     83.38
229.04
         286.30
                 83.38
                          286.30
                                   286.30 83.38
-286.30
         229.04
                 83.38
                         -229.04
                                   229.04 83.38 -171.78
                                                           229.04
                                                                    83.38
                                  229.04 83.38
229.04 83.38
                 83.38
                         -57.26
                                                    0. 229.04 83.38
-114.52
         229.04
                                                  171.78
  57.26
         229.04
                 83.38
                          114.52
                                                           229.04
                                                                    83.38
                 83.38
                                   229.04 83.38
229.04
         229.04
                          286.30
                                   171.78 83.38 -171.78
-286.30
         171.78
                 83.38
                         -229.04
                                                           171.78
                                                                    83.38
-114.52
         171.78
                 83.38
                         -57.26
                                   171.78 83.38
                                                   0. 171.78 83.38
 57.26
         171.78
                 83.38
                          114.52
                                   171.78 83.38
                                                   171.78 171.78
                                                                    83.38
         171.78
                                   171.78 83.38
229.04
                 83.38
                          286.30
-286.30
         114.52
                 83.38
                         -229.04
                                   114.52 83.38 -171.78
                                                           114.52
                                                                    83.38
-114.52
         114.52
                 83.38
                        -57.26
                                   114.52 83.38
                                                    0. 114.52 83.38
  57.26
         114.52
                 83.38
                         114.52
                                   114.52 83.38
                                                   171.78
                                                           114.52
                                                                     83.38
229.04
         114.52 83.38
                          286.30
                                   114.52 83.38
-286.30
         57.26
                 83.38
                        -229.04
                                   57.26 83.38 -171.78
                                                            57.26
                                                                    83.38
                                                   0. 57.26 83.38
                          -57.26
                                   57.26 83.38
-114.52
         57.26
                 83.38
 57.26
         57.26
                 83.38
                          114.52
                                    57.26 83.38
                                                   171.78
                                                             57.26
                                    57.26 83.38
229.04
         57.26
                 83.38
                          286.30
-286.30
         0.83.38
                   -229.04
                               0. 83.38 -171.78
                                                     0.83.38
                    -57.26
-114.52
         0.83.38
                                0. 83.38
                                            0. 0. 83.38
                    114.52
286.30
                                           171.78
 57.26
         0.83.38
                                0. 83.38
                                                      0.83.38
229.04
         0. 83.38
                                0.83.38
-286.30
        -57.26
                 83.38
                        -229.04
                                 -57.26
                                          83.38 -171.78
                                                         -57.26
-114.52
        -57.26
                 83.38
                         -57.26
                                  -57.26
                                          83.38
                                                    0. -57.26 83.38
                                                  171.78 -57.26
 57.26
        -57.26
                 83.38
                          114.52
                                  -57.26
                                          83.38
                                                                    83.38
        -57.26
                 83.38
                          286.30
                                  -57.26
                                          83.38
229.04
        -114.52
                 83.38
                         -229.04
                                 -114.52
                                                 -171.78
-286.30
                                          83.38
                                                         -114.52 83.38
        -114.52
-114.52
                 83.38
                         -57.26
                                 -114.52
                                          83.38
                                                    0. -114.52 83.38
        -114.52
                                          83.38
  57.26
                 83.38
                          114.52
                                 -114.52
                                                  171.78
                                                          -114.52 83.38
        -114.52
                          286.30
                                 -114.52
229.04
                 83.38
                                          83.38
-286.30
        -114.52
                 83.38
                         -229.04
                                 -171.78
                                          83.38
                                                 -171.78
                                                          -171.78 83.38
                         -57.26 -171.78
                                                    0. -114.52 83.38
-114.52
        -171.78
                 83.38
                                          83.38
                                                  171.78 -171.78 83.38
 57.26
        -171.78
                 83.38
                          114.52 -171.78
                                          83.38
                          286.30 -171.78
229.04
        -171.78
                 83.38
                                          83.38
-286.30
        -114.52
                 83.38
                         -229.04 -229.04
                                          83.38
                                                 -171.78
                                                           -229.04 83.38
-114.52
        -229.04
                 83.38
                         -57.26 -229.04
                                          83.38
                                                    0. -114.52 83.38
        -229.04
                 83.38
  57.26
                          114.52 -229.04
                                          83.38
                                                  171.78
                                                          -229.04 83.38
229.04
        -229.04
                 83.38
                          286.30 -229.04
                                          83.38
                 83.38
        -114.52
                         -229.04 -286.30
                                          83.38
                                                 -171.78
-286.30
                                                         -286.30 83.38
        -286.30
                          -57.26
-114.52
                 83.38
                                 -286.30
                                          83.38
                                                   0. -114.52 83.38
 57.26
        -286.30
                          114.52 -286.30
                                                  171.78 -286.30 83.38
                 83.38
                                          83.38
229.04
        -286.30
                 83.38
                          286.30 -286.30
                                          83.38
```